

17th DOE NUCLEAR AIR CLEANING CONFERENCE

Session 7

FILTRATION AND FILTER TESTING II

WEDNESDAY: August 4, 1982
CO-CHAIRMEN: H. Gilbert
Consultant
F.J. Freibert
Rockwell International

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REPORT OF MINUTES OF GOVERNMENT-INDUSTRY MEETING ON FILTERS, MEDIA, AND MEDIA TESTING

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A well-informed group dedicated to solving the problems of high efficiency filters and the methodology of their evaluation has been in existence for many years. The contributions of this group have spanned the last ten air cleaning conferences. From the original individuals meeting in a hotel room in a late hour rump session format, the sessions have evolved into a formal group with a permanent chairman and a prepared agenda. The original open and often argumentative mode of operation has been replaced with professional technical papers based upon original and/or continuing research programs. This format allows a communication exchange at the working level and a platform for the early presentation of experimental data on technical topics.

The most recent session of this working group was held on Monday, August 2, 1982, just preceding the 17th DOE Nuclear Air Cleaning Conference in Denver, Colorado. Over 250 attendees participated in the discussions of current interest. A wide spectrum of groups were represented with foreign attendance greater than any previous conference. This session, following the precedents of earlier meetings, related the operating requirements placed on the high efficiency filter and the capacity of industry to meet them. To this end, the collected talents of the assembled body were unified toward the problems of the particulate filter, its components, and its method of test. Representatives of all facets of the industrial world were present, from the basic fiber suppliers, through media producers, and finally to the filter unit fabricators. Research organizations, academic institutions and foreign visitors all contributed to the discussions.

The following review contains an executive overview of the items of deliberation. The subjects are addressed in abstract form and are listed in their order of presentation and not in priority. Six separate lissues were discussed; complete papers are attached as appendices to this overview.

Review of DOE Filter Test Facilities (1970-1980)

Cliff Burchsted from ORNL summarized the operation of the three DOE filter test stations over the 1970-80 time span. Included were the totals of the units tested, together with incremental test station breakout for both DOE and non-DOE customers. Total rejection experiences were also reported with specific separation of the data reflecting penetration and quality assurance as major contributors. A number of conclusions were reported: DOE testing requirements were increasing and non-DOE decreasing; total testing was constant but over the years showed cycling which was associated with new facility startup; rejections resulting from excessive penetration were constant over the time span, and quality assurance

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failures have increased over the recent years. This latter conclusion has been a concern of DOE and will result in an NRC reexamination of its inspection criteria for required testing. The complete documentation is attached as Appendix A.

Recent DOE Filter Test Operations

Jim Bresson from DOE (Albuquerque) addressed several concerns that resulted from the previous San Diego meeting and reported to the group the interim actions that have been accomplished. Simply stated, these concerns are (1) the physiological safety of DOP as a test aerosol and (2) the loss of credibility of the test stations in the evaluation process. An in-house standards writing group has been establishing specific documentation to clearly define test station policies, operating procedures, and finally filter unit specifications. The first draft of these documents has been internally reviewed, and suggested revisions incorporated. The resultant document has been distributed to a peer group of technical experts and industrial users for comprehensive review and critique. Comments from this group will be addressed and after resolution will be incorporated into the final document. Technical merit will be an overall consideration before decision and implementation will be recommended. As a part of the policy developed, consideration will be given for procedures necessary for the qualification of additional test aerosols and/or new devices/techniques for aerosol quantification. Two additional statements by Bresson to the assemblage were important: (1) Los Alamos laboratory will provide technical support to the test facilities, and (2) round robin testing will be implemented by the test stations to assure uniformity of procedures and results. Further comments by Jim Bresson are contained in Appendix B.

Survey of HEPA Filter Experience

Carbaugh from Pacific Northwest Laboratory (Batelle, Richland) summarized the results of a survey of HEPA filter applications to provide insight into the reasons for and magnitude of filter changeovers and failures. Included were some interesting statistics that 58% of the units surveyed were changed and that 18% changed more than once. Most of the changes (63%) were due to excessive pressure drop followed by leak test failures (15%). Other conclusions reached were:

1. Filters performed their designed tasks.
2. Changes resulted from performance degradation rather than failure.
3. Accurate records are not generally available to determine failure causes.
4. Chemical and other environmental conditions were contributing factors for failure.

Associated data and additional comments are available in Appendix C.

Aerosol Size Distribution at Filter Test Stations

Gary Salzman from Los Alamos laboratory elaborated on a portion of the test support of the field stations mentioned earlier by Jim Bresson. As a part of their continuing effort to replace/supplement the instrumentation for aerosol size measurement and quantification, they have obtained a commercial, single particle, laser spectrometer for further study. The initial effort has concentrated on the measurement of particle size and size distribution at the test stations. They have developed microprocessor technology for data accumulation, interpretation, and display in real time sequences. Presently these data are confined to size and size distribution functions. Data obtained thus far by this instrument show a considerably smaller average size and a much wider size distribution than other devices used previously. They have also concluded that the instrument is relatively insensitive to changes in aerosol concentration and refractive index over the range of interest for filter testing. These conclusions stimulated a wide variety of questions and the discussions that followed were of a highly technical nature with little resolution of the apparent differences in stated opinions. With this in mind, the paper should be considered as a progress report, and we will be looking forward to additional concepts/recommendations as the LASL support to the test stations continues. Further amplification and data presentation are given in Appendix D.

DEHS as a Challenge Aerosol in Filter Testing

Julie McIntyre from the Hanford Environmental Health Foundation (Richland) reviewed their ongoing efforts where the hexyl sebacate (DEHS) has been used as a challenge aerosol substitute for the hexyl phthalate (DOP). Characterization of the DEHS aerosol as well as operating conditions were briefly described. The magnitude of the data presented will require additional analysis beyond this meeting to digest and interpret. As a result of their progress to date, Hanford has reached the following conclusions: DEHS has been found to be compatible with existing test station facilities and can effectively discriminate between acceptance/rejection penetration performance levels; testing shows compatibility between DOP and DEHS. They finally concluded that DEHS is an acceptable challenge aerosol in HEPA filter testing. Complete data and specific comparative analyses for this conclusion are given in Appendix E.

Need for NRC Regulatory Code Revisions

Regulatory Guide 1.52, Design, Testing and Maintenance Criteria for Nuclear Power Plants, was last revised in 1978. Recent events and data generated during the Three-Mile Island incident have dictated that the guide should be reexamined and issued in a revised form. Dick Bangart from NRC outlined the factors that have contributed to the need for the revision and those elements that influence the timetable for revision. Among those stated are: NRC endorsement of ANSI 509 and 510 must be accomplished; NRC must review its present position of not requiring HEPA filter testing at DOE test facilities; and post-TMI, accident-related concerns must be addressed in the

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light of new research data on adsorbent weathering from the Naval Research Laboratory. It was stated that efforts to revise the guide will be made in FY 83. Industry was requested to identify candidate issues and supply support data for incorporation into the final document. It is anticipated that the proposed revision will continue to endorse consensus standards developed by air cleaning technical experts. The complete text is attached as Appendix F.

Only one issue in the area of new business was raised from the floor. This was stated in the form of an appeal by Mr. Jacox for assistance in the examination/revision of ANSI 509 and 510. He stated that technical expertise was urgently needed; general subject areas with their specific points of contact were announced for those interested in further participation.

In conclusion, it should be reemphasized that this informal working group, with its diversified representation, provides a means for a comprehensive and expedient solution to the problems of the high efficiency filtration industry. The total effort has proven invaluable because it permits the surfacing and exposure of problems that might otherwise be lost in the quagmire of bureaucracy and management. The meetings are intended to be, and actually are, a working level distribution of data and expertise as well as a progress report of ongoing projects in the particle filtration areas. To this end, the group feels they have been successful and future sessions are contemplated.

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Appendix A

A REVIEW OF DOE FILTER TEST FACILITY OPERATIONS* 1970-1980

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INTRODUCTION

Filter test facilities (FTFs) are maintained for the U.S. Department of Energy (DOE) at Oak Ridge, Tennessee (Union Carbide Corporation Nuclear Division); Rocky Flats, Colorado (Rockwell International); and Richland, Washington (Hanford Environmental Health Foundation). The function of these FTFs (or stations, as they are usually called) is to verify critical performance characteristics (penetration and airflow resistance) and compliance-with- specifications of high efficiency particulate air (HEPA) filters used in critical air and gas cleaning systems of DOE facilities. The stations test and examine each filter individually; acceptance on the basis of statistical sampling is not permitted. The stations also, on a contract basis, test and inspect filters for other government agencies and for commercial organizations on request.

At the DOE's Airborne Waste Management (AWM) workshop at San Francisco on March 6, 1981, it was requested that the operating experience of the FTFs for the past several years be reviewed. This report presents the findings and conclusions of that review.

Test Load

Table 1** and Fig. 1 show the number of filters tested by each station and the total number of filters tested by all stations for each year since 1970. It can be seen from Fig. 1-a that, if the 1972 and 1976 peaks are ignored (both occurred at Hanford and reflect major filter replacement programs at certain DOE facilities), there has been a slow but steady increase in the total number of filters tested in the program (a trend line ignoring these peaks would have a positive slope of about 200 filters per year). As can be seen from Fig. 1-b through 1-d, however, there has been a major redistribution of test loads since 1976. This redistribution is seen more clearly in Fig. 2. The Hanford (HF) station after peaking in 1976 shows a steady downward trend since that time. The trend at the Oak Ridge (OR) station shows a steady decline over the entire period. The pronounced change came with the startup of the Rocky Flats (RF) station in 1974 (reports from RF were not available until 1976). Since 1976, the RF share of the test load has increased from 15.7% to 41.1% of the total tested, while OR and HF shares have dropped to 34.5% and 24.4%, respectively. An additional part of the FTF load comes from testing of gas-mask canisters. This experience is shown in Table 1-a; it is included for information only and is not pertinent to the report.

*Research sponsored by the Defense Waste and Byproducts Division, U.S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

**Tables are placed at the end of the report.

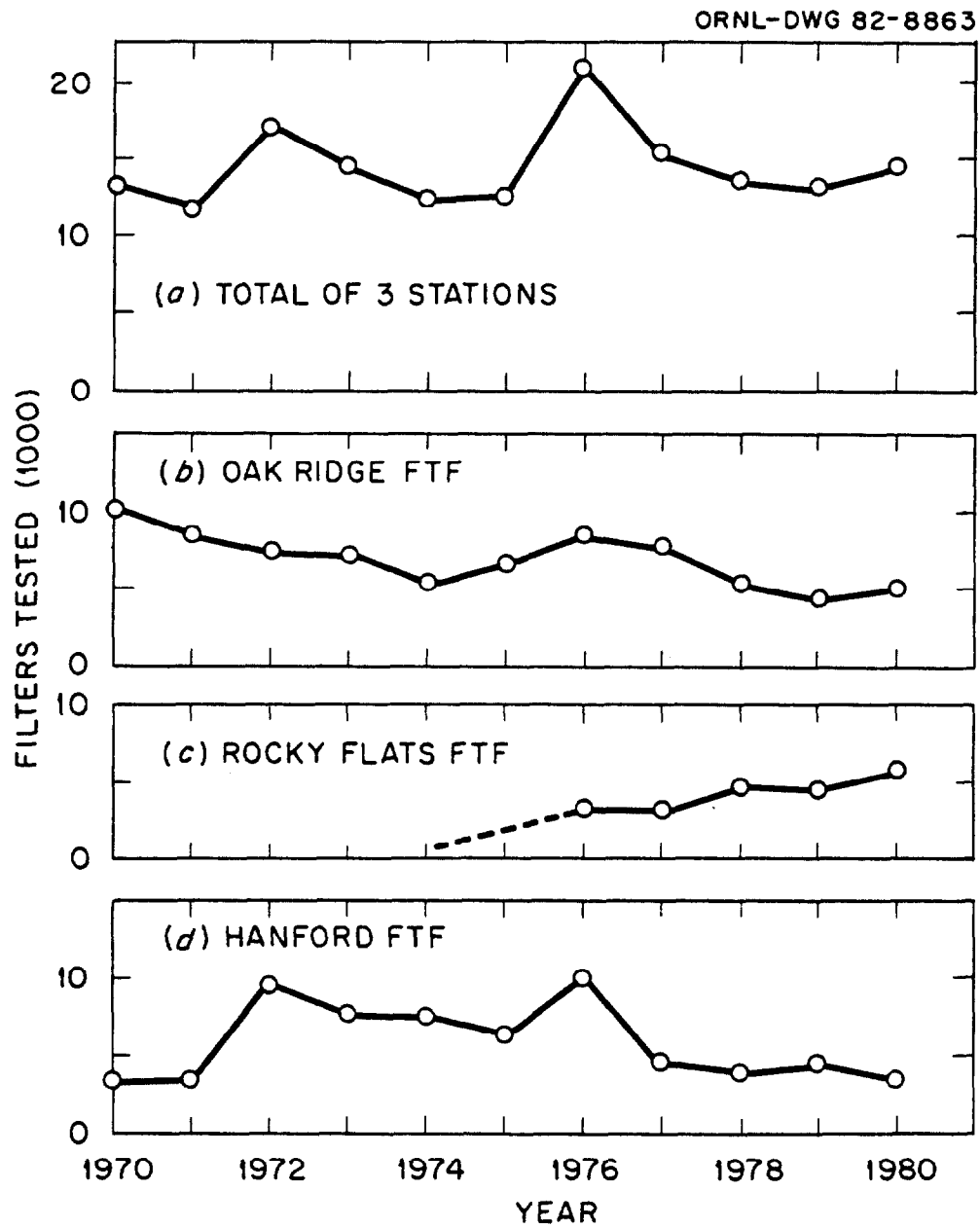


Fig. 1. Number of HEPA Filters tested by DOE Filter Test Stations, January 1970-March 1980.

ORNL-DWG 82-8867

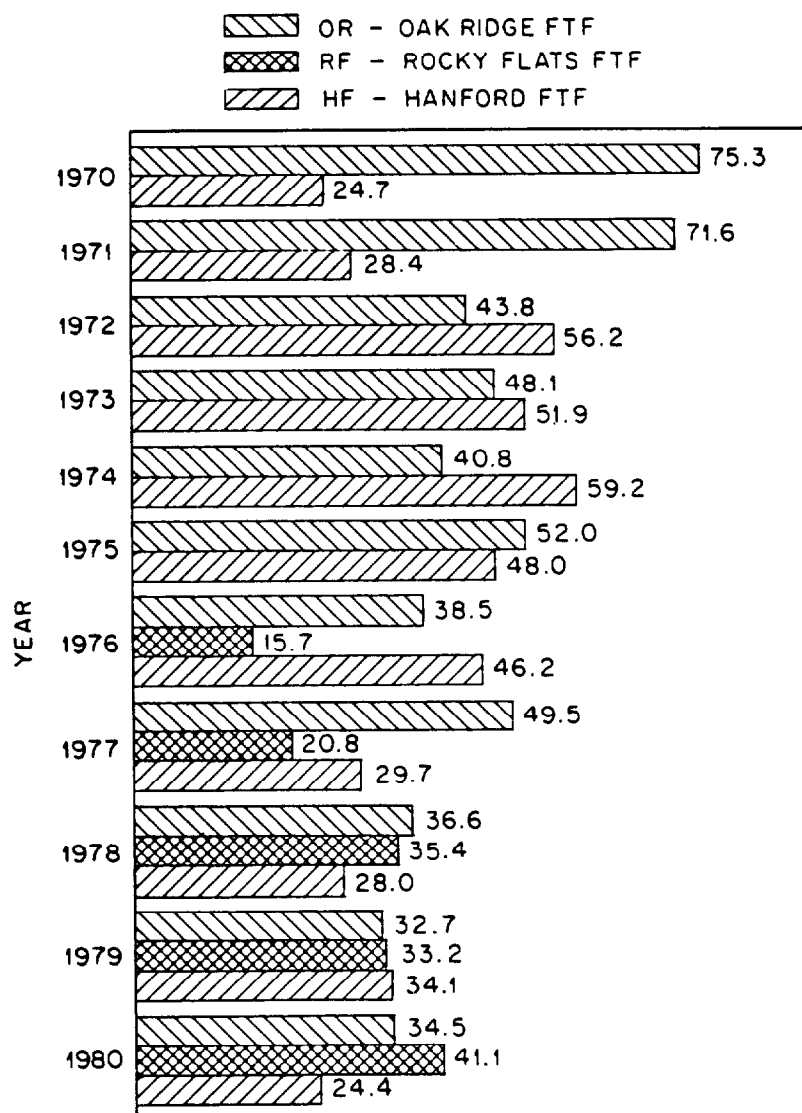


Fig. 2. Percent of total filters tested by each Filter Test Stations, January 1970-March 1980

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Distribution of Test Load by Customer

FTF testing is considered so important by DOE that the service is provided to its contractors at no direct charge. Because the stations have excess capacity, the service is also provided to other government agencies and to commercial organizations at cost. Table 2 and Fig. 3 shows the distribution of filters tested for DOE contractors and for non-DOE customers. Fig. 3 shows that, although the number of filters tested for DOE contractors has increased (this can be seen more clearly if the 1972 and 1976 HF peaks are ignored). The number of filters tested for non-DOE customers, after reaching a peak in 1977, has fallen sharply. We suspect that this drop was in large part due to the Nuclear Regulatory Commission (NRC) decision to cease recommending FTF verification of HEPA filters purchased for commercial nuclear power stations. NRC regulatory guide (RG) 1.52 had previously recommended FTF testing of filters intended for use in commercial nuclear power plants.² At the 15th DOE Nuclear Air Cleaning Conference (Boston, Massachusetts, August 1978), J. T. Collins announced that, as the result of a review of FTF operational data made by the NRC staff, NRC would no longer recommend verification testing of filters.³ The result of this pronouncement is seen in the decrease in non-DOE customer testing since 1978 shown by Fig. 3-b. In addition, the Navy Ships Parts Supply Center announced shortly after the pronouncement that they also would no longer require FTF testing. Discussions between FTF personnel and filter manufacturers indicated that most utilities had opted against further FTF testing as a result of the NRC position, although the major manufacturers still believe it is needed. Fig. 4, which shows non-DOE customer testing as a percent of total test load, shows this drop in non-DOE customer testing even more clearly. The NRC action will be discussed further in the next section of this report. In closing this section, however, it should be noted that a considerable number of filters continue to be tested for non-DOE customers, including apparently, some for commercial nuclear power stations.

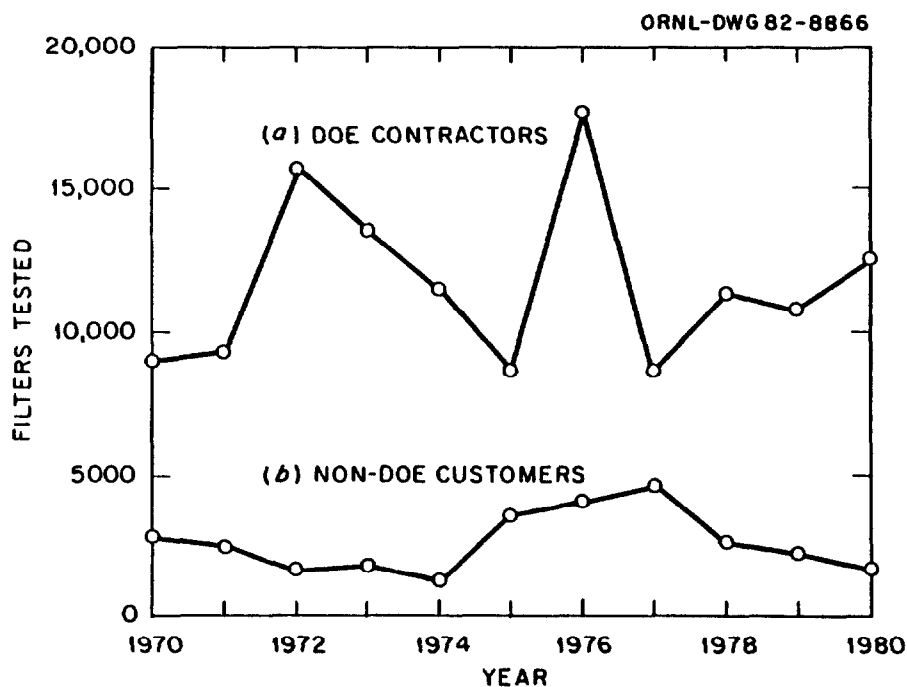


Fig. 3. Distribution of FTS test load between DOE contractors and non-DOE customers, 1970-1980

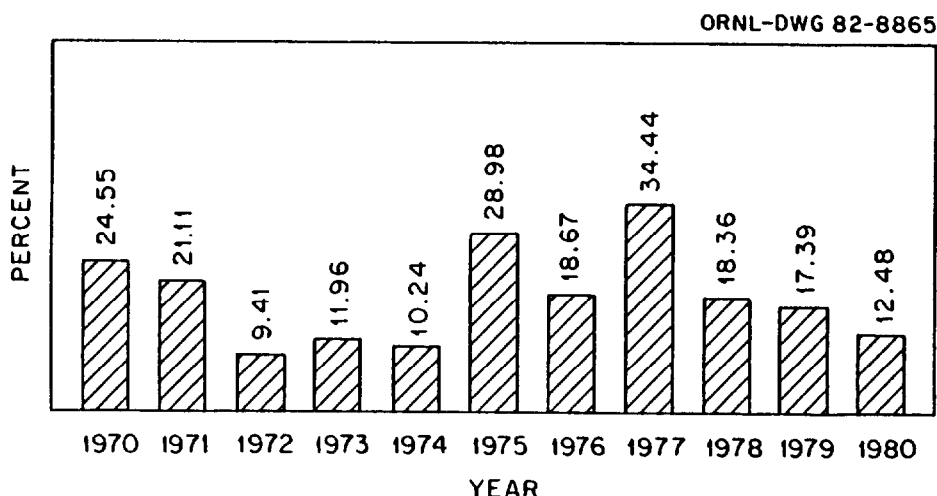


Fig. 4. Filters tested for non-DOE customers as a percent of total filters tested, 1970-1980

Rejection Experience

Tables 3 and 4 list the number of filters rejected for failure to meet penetration requirements and failure to meet other specification requirements, respectively. The data of these tables are shown as percentages of the number of filters tested in Tables 5 and 6 respectively.

It can be seen from Fig. 5 that, except for one particularly bad year in 1973, the rejection of HEPA filters for excessive penetration has not varied greatly from year to year, averaging 2.03% for the period of the report. There is really no trend in such rejections either up or down; this is to be expected. Penetration is primarily a function of filter medium (i.e., paper) quality and basic filter design. Filter manufacturers purchase their paper to very rigid specifications,⁶ and seal the "pack" made from it by a carefully qualified procedure. Unless the paper is damaged in the course of manufacture or handling or unless the packsealing procedure is compromised during manufacture, one would not expect to see any great amount of filter rejection for penetration. The 1973 peak was probably an anomaly and the condition reflected by it was rectified through consultation of DOE personnel with the manufacturer(s) involved.⁷

Filter rejection for workmanship and other causes related to quality assurance (QA) during manufacture and shipping is another story. QA-related rejections, which reflect day-to-day activities of the filter manufacturer, include excessive pressure drop; failure to meet certain specification requirements with respect to design or construction; mislocated or loose gaskets; physical damage; and shipping damage. High pressure drop, though related to filter medium quality, is basically a function of the area of paper in the filter pack and can vary somewhat from filter to filter. Shipping damage is included because it most often stems from inadequate package design or inadequate shipping procedures. Rejection history of the Hanford station for QA-related causes is shown in Fig. 6. Hanford data are used in this figure because they are the most meaningful for this purpose. Hanford reports all rejectable filters and

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meaningful for this purpose. Hanford reports all rejectable filters and does no repair.⁸ Rocky Flats station, on the other hand, repairs as many filters as it can; often waives what they consider non-critical defects or noncompliances; and does not report repaired filters as rejects.⁹ Oak Ridge practice lies somewhere between these extremes.¹⁰

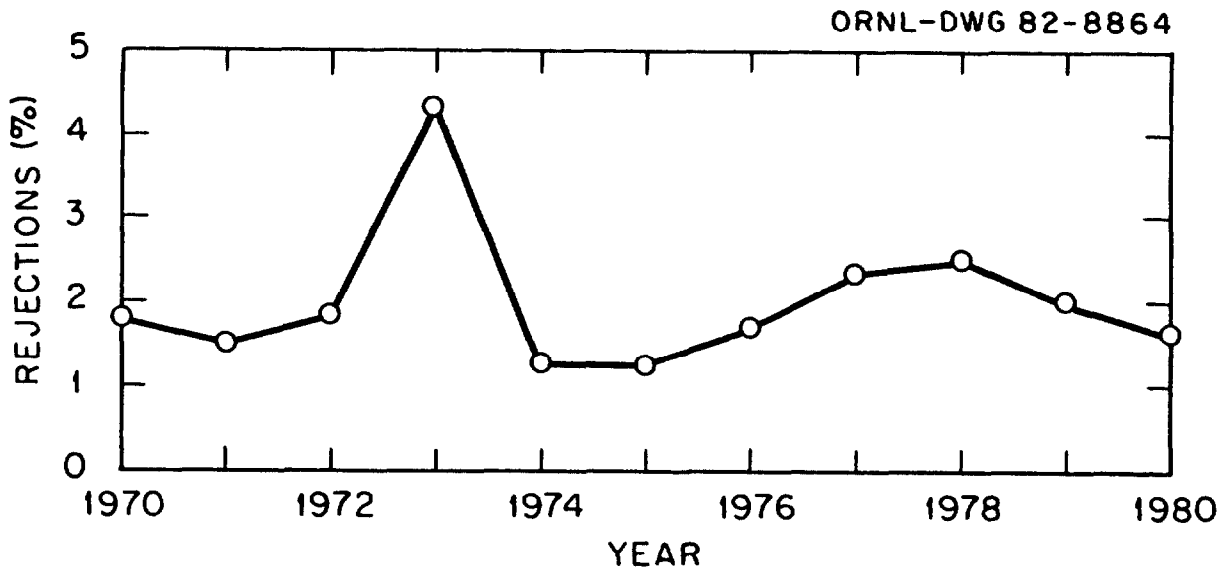


Fig. 5. HEPA filter rejections for penetration, as a percent of total filters tested, 1970-1980.

As can be seen from Fig. 6, which is believed to reflect the true rejectable filter situation, QA-related rejection has been somewhat cyclic, peaking and falling off periodically as apparently manufacturers alternately relax and tighten up their QA practices. The average rejection rate for QA-related deficiencies for the period 1970 through 1977 was 4.6%, but has climbed strikingly since 1978. The 1979 and 1980 rates, 12.14% and 26.75% respectively, are both significantly higher than those for any preceding year during the period of this report.

It is believed that this increase in QA-related rejection may stem in part from the NRC action of 1978 and represents a possibly significant decrease in filter quality assurance. It is, perhaps, significant that the increase started in the second half of 1978 following the issue of revised RG 1.52 in March of that year. At the end of Collins' presentation at the Air Cleaning Conference (August 1978), an officer of one of the major HEPA filter manufacturers commented that he believed that the proposed relaxation of FTF testing could lead to a deterioration of filter quality, perhaps to the development of two distinct quality levels for filters.¹¹ This increase in QA-related rejections appears to reflect that postulated deterioration of quality. This is a development that must be watched closely and which should be a major topic for discussion at future AWM-FTF workshops and meetings of the Government-Industry Conference on Filters and Filter Media.⁷ To show that the increase in QA-related rejections is not peculiar to Hanford, the total QA-related rejections for all three stations are shown in Fig. 7; the same increase is evidenced in Fig. 7, although it is obvious that Hanford is the major influence on the curve. It is reasonable to assume that, since manufacturers are aware that the filters will be carefully inspected and tested at the FTF, that the

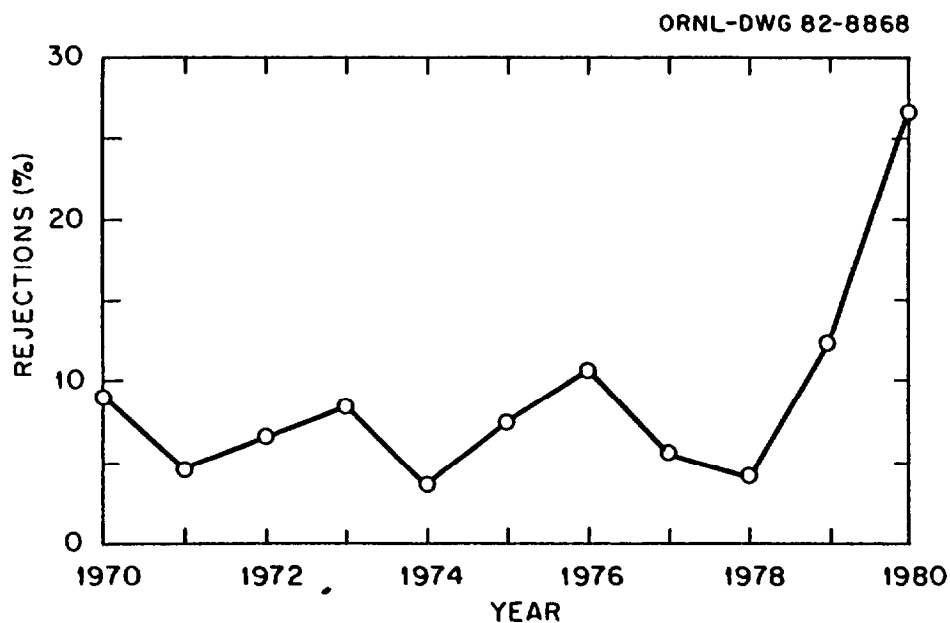


Fig. 6. QA-related rejections at Hanford FTS as a percent of total filters tested at that stations, 1970-1980

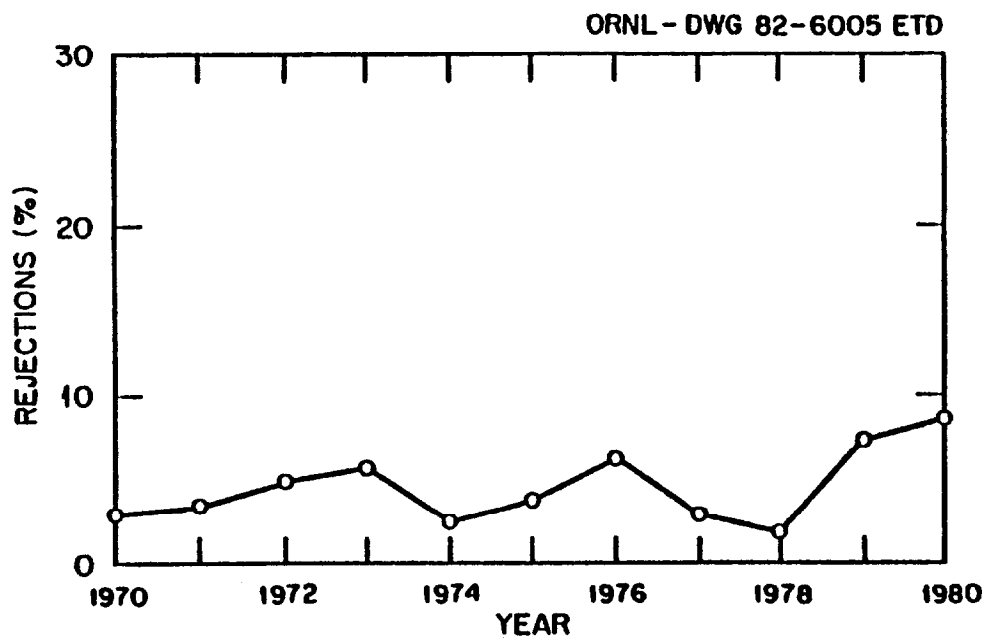


Fig. 7. QA-related rejections for all stations as a percent of total filters tested, 1970-1980

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DOE is receiving the "cream of the crop" of HEPA filters. Considering the wide swings in rejection rate for quality-assurance-related factors still seen in this program, there is some question concerning the quality of HEPA filters being sent to users who do not avail themselves of the FTF service.

Differences Between Station Operations

Fig. 1 illustrated the differences and changes in test loads of the three stations and the previous section discussed major differences between the stations with respect to rejection and repair policy. Another major difference between the stations is in their report policies. The operating reports vary widely in the amount of detail provided and these differences make analyses of the type described in this report difficult. The deviation in reporting practices is particularly evident in the data of Table 7. This table was prepared as the basis for a comparison of rejection rates for penetration in which bias was minimized by showing data for only a single size of filter made by a single manufacturer. The analysis could not be made because of the failure of Rocky Flats to provide any such information and the failure of Hanford to supply such information after 1972. Greater uniformity in reporting would greatly simplify the periodic reviews of station operations and permit better comparison of policies and practices.

Conclusions

1. The total test load for the three stations is increasing.
2. The number of filters tested for DOE contractors is increasing.
3. The number of filters tested per year for non-DOE customers has dropped substantially since 1978, probably as a result of the NRC decision to no longer recommend FTF testing of filters for commercial nuclear power plants.
4. The NRC pronouncement of filter testing has manifestly had an adverse affect on the number of filters received for test since 1978.
5. The test loads at Oak Ridge and Hanford are decreasing. Conversely, the test load of Rocky Flats is increasing.
6. Significant differences in inspection, repair, and acceptance policies exist between the three stations.
7. Significant and undesirable differences in report format and content exist between the three stations.
8. Test machine operating practices are probably consistent from station to station, but have varied in the past. Periodic round-robins are needed to maintain and verify consistency of testing.
9. The continued operation of the test stations is essential to maintain minimum levels of HEPA filter quality for nuclear applications.

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10. The NRC should be notified of the findings of this review.
11. Wide swings in rejection rates for quality-assurance-related factors still continue. Since DOE probably receives the cream of the crop of the HEPA filters made, this leaves some question concerning the quality of filters used by organizations that do not utilize FTF inspection and testing.

NOTES AND REFERENCES

1. Semiannual reports of the FTFs were combined to show annual figures. The data for 1977 were adjusted for the change from a July-June to an October-September fiscal year.
2. U. S. Nuclear Regulatory Guide 1.52, *Design, Testing, and Maintenance Criteria for Engineered-Safety-Feature Atmosphere Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants*.
3. J. T. Collins, R. R. Bellamy, and J. R. Allen, *Evaluation of Data from HEPA Filter Quality Assurance Testing Stations*, PROCEEDINGS OF THE 15th DOE NUCLEAR AIR CLEANING CONFERENCE, 7-10 August 1978.
4. Personal communication, Anthony Alexander, Navy Ships Part Supply Center, to C. A. Burchsted.
5. Personal communications, Managers of DOE filter test stations to filter manufacturers as reported to C. A. Burchsted.
6. Military Specification MIL-F-51079, *Filter Medium Fire-Resistant High Efficiency*.
7. Filter manufacturers and responsible government personnel meet regularly to discuss industry and user problems at the Government-Industry Conferences held in conjunction with the biennial DOE Nuclear Air Cleaning Conferences.
8. Personal communication, R. Gilmore, Manager of Hanford FTF, to C. A. Burchsted.
9. Personal communication, W. E. Elliott, Manager of Rocky Flats FTF, to C. A. Burchsted.
10. Personal communication, R. C. May, Manager of Oak Ridge FTF, to C. A. Burchsted.
11. Response of George Caldwell, Vice President of Flanders Filters, Inc., to paper of Reference 3, PROCEEDINGS OF 15th DOE NUCLEAR AIR CLEANING CONFERENCE, 7-10 August 1978.

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Table 1

Number of Filters Tested by FTFs, 1970-1980

Year	Oak Ridge	Rocky Flats	Hanford	Total
1970	9,939	--	3,263	13,202
1971	8,540	--	3,388	11,928
1972	7,506	--	9,645	17,151
1973	7,165	--	7,730	14,895
1974	5,217	NR*	7,570	12,787
1975	6,631	NR*	6,129	12,760
1976	8,340	3,399	9,987	21,626
1977	7,740	3,257	4,640	15,637
1978	5,118	4,945	3,914	13,977
1979	4,311	4,368	4,497	13,176
1980	4,966	5,909	3,507	14,382
Totals	75,473	21,878	64,270	161,521
Yearly Average	6,681	4,376	5,843	14,684

*NR = No report issued for initial years of operation.

Table 1-a

Number of Gas-Mask Cartridges Tested by FTFs, 1970-1980

Year	Oak Ridge	Rocky Flats	Hanford	Total
1970	5,573	--	39,486	45,059
1971	4,486	--	14,977	19,463
1972	3,545	--	5,669	9,214
1973	3,361	--	3,999	7,360
1974	3,677	--	11,181	14,858
1975	5,696	--	5,228	10,924
1976	9,484	No Data	5,491	14,975
1977	16,132	No Data	7,077	23,209
1978	15,250	No Data	14,181	29,431
1979	31,627	No Data	23,546	55,173
1980	17,643	No Data	36,729	54,372
Totals	116,474	No Data	167,564	284,038

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Table 2

Filters Tested By Customer and FTF, 1970-1980

Year	Oak Ridge		Rocky Flats*		Hanford		Totals	
	DOE	Non-DOE	DOE	Non-DOE	DOE	Non-DOE	DOE	Non-DOE
1970	6,303	2,544	--	--	2,681	379	8,984	2,923
1971	6,369	2,135	--	--	3,013	375	9,382	2,510
1972	6,362	1,128	--	--	9,297	498	15,659	1,626
1973	5,818	1,371	--	--	7,722	468	13,540	1,839
1974	4,029	1,188	NR	NR	7,449	121	11,478	1,309
1975	3,305	2,967	NR	NR	5,507	629	8,812	3,596
1976	4,839	3,485	3,229	170	9,508	379	17,576	4,034
1977	2,580	3,970	2,931	326	3,249	297	8,760	4,601
1978	3,278	1,840	4,327	618	3,806	108	11,411	2,566
1979	3,523	788	3,931	437	3,430	1,066	10,884	2,291
1980	3,887	999	5,377	532	3,253	254	12,517	1,785
Totals	50,293	22,423	19,795	2,083	58,915	4,574	129,003	29,080
Yearly Average	4,572	2,038	3,959	417	5,356	416	11,728	2,644

*Rocky Flats' figures are estimates based on percentages provided by W. E. Elliott, Manager of Rocky Flats

Table 3

Number of Filters Rejected for Excessive Penetration, 1970-1980

Year	Oak Ridge	Rocky Flats	Hanford	Total
1970	149	--	90	239
1971	70	--	98	168
1972	209	--	139	308
1973	266	--	379	645
1974	67	NR	95	162
1975	30	NR	129	159
1976	194	53	121	368
1977	63	193	108	364
1978	137	167	46	350
1979	82	92	97	271
1980	68	74	97	239
Totals	1,335	579	1,399	3,273
Yearly Average	121	116	127	298

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Table 4

Number of Filters Rejected for QA-Related Causes, 1970-1980

Year	Oak Ridge	Rocky Flats	Hanford	Total
1970	106	--	292	398
1971	236	--	161	397
1972	170	--	651	821
1973	213	--	642	855
1974	32	NR	288	320
1975	61	NR	468	529
1976	127	151	1,084	1,352
1977	58	121	270	449
1978	74	26	162	262
1979	379	32	546	957
1980	278	18	938	1,234
Totals	1,734	348	5,502	7,574
Yearly Average	158	70	500	688

Table 5

Filters Rejected for Penetration, Percent of Total Tested
1970-1980

Year	Oak Ridge (%)	Rocky Flats (%)	Hanford (%)	Total (%)
1970	1.50	--	2.76	1.81
1971	0.82	--	2.89	1.41
1972	2.78	--	1.44	1.80
1973	3.71	--	4.90	4.33
1974	1.28	NR	1.25	1.27
1975	0.45	NR	2.10	1.25
1976	2.33	1.56	1.21	1.70
1977	0.81	5.93	2.33	2.33
1978	2.68	3.38	1.18	2.50
1979	1.90	2.11	2.16	2.06
1980	1.37	1.25	2.77	1.66
Average	1.77	2.65	2.18	2.03

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Table 6

Filters Rejected for QA-Related Deficiencies, Percent of
Total Tested, 1970-1980

Year	Oak Ridge (%)	Rocky Flats (%)	Hanford (%)	Total (%)
1970	1.07	--	8.95	3.01
1971	2.76	--	4.75	3.33
1972	2.26	--	6.75	4.79
1973	2.97	--	8.31	5.74
1974	0.61	NR	3.80	2.50
1975	0.92	NR	7.64	3.67
1976	1.52	4.44	10.85	6.25
1977	0.75	3.72	5.82	2.87
1978	1.45	0.53	4.14	1.87
1979	8.79	0.71	12.14	7.26
1980	5.60	0.30	26.75	8.58
Average	2.30	1.59	8.56	4.69

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Appendix B DOE FILTER TEST PROGRAM

POLICY FOR THE '80s

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During the 1978-1980 time period, two issues related to the Department of Energy (DOE) HEPA filter test activities were raised. First was the safety issue of continued use of DOP as the authorized test aerosol, since there was some indication that the material might be classified as a carcinogen. As a result, one of the three DOE filter test facilities stopped using DOP as the test aerosol, a prohibition which continues. Second and harder to define, were rumors that the filter test facilities were not producing uniform test results, and that filters rejected by one facility might pass at another. Although the source and accuracy of these rumors was never truly identified, it was determined that the round robin tests, once conducted periodically by all three facilities, had not been performed in at least three years.

Based on the above information, DOE, through the joint efforts of the Airborne Waste Management Program Office at the Idaho Operations Office, and the Interim Waste Operations Program Office at DOE Headquarters, Germantown, MD, made the following decisions:

1. Continue to support filter test activities at the three DOE filter test facilities.
2. Establish a formal policy on filter testing and procedural requirements for the test operations performed at the filter test facilities.
3. Establish a standards writing group to write the DOE policy statement and procedural requirements.
4. Establish a plan for technical support.
5. Restore round robin testing.

The Standards Writing Group was established in March 1981 and has to date:

1. Completed a draft policy statement on testing of filters to be used at DOE facilities.
2. Completed a draft set of procedural requirements to be implemented at each filter test facility.
3. Completed a first draft quality program plan, under which site-specific QA/QC procedures must be written.

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4. Discussed, but not yet written, manufacturing specifications for filters to be used at DOE facilities.

Draft documents identified in items 1 and 2 above have been sent to a carefully selected peer review group which consists of DOE and non-DOE related technical experts. In the latter category are representatives from filter and test instrument manufacturers.

The proposed procedures contain two very important considerations. The first consideration is a mechanism whereby, under strict requirements, aerosols other than DOP can be evaluated, tested, and authorized for use in DOE filter test facilities. The requirements include toxicology and other health and safety concerns such as fire and explosion. The second consideration is a mechanism whereby new measurement equipment or techniques can be tested and authorized for use in DOE filter test facilities. Thus, test methods and aerosols which can be shown to reliably produce filter test results equivalent to those obtained by the current method, can be approved for use in the DOE filter test facilities. DOE is not considering replacing DOP or currently used test equipment at this time, but wants to have the capability to make changes in case of need or desirability.

DOE is committed to the following policy as the HEPA Filter Test Program is developed and implemented. First, the technical merits of any change in current test methods, including the aerosol used for test, must be demonstrated. Second, the proposed changes must be subject to a rigorous peer review. Third, issues raised by peer review must be satisfactorily resolved prior to approval of proposed changes.

One of the items mentioned earlier in this talk referred to technical support. The DOE Airborne Waste Management Program Office at the Idaho Operations Office has funded a group from the Los Alamos National Laboratory (LANL) to provide this service. This group's first task is to evaluate the suitability of a single particle analyzer to measure particle size and size distribution of test aerosols. A paper on this work is to be presented later in this session. Next fiscal year, the LANL group will begin work on identifying possible alternatives to DOP. In addition, they will provide technical support to filter test facilities as required. In this past year, the LANL people have helped install, and make functional, single particle analyzers and associated dilution boards at each of the three filter test facilities.

I hope this short presentation provides sufficient information to allay some of the concerns you have had with respect to DOE HEPA filter test activities. I would be happy to answer any questions you may have on these matters.

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Appendix C

A SURVEY OF HEPA FILTER EXPERIENCE*

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Abstract

A survey of high efficiency particulate air (HEPA) filter applications and experience at Department of Energy (DOE) sites was conducted to provide an overview of the reasons and magnitude of HEPA filter changeouts and failures. Results indicated that approximately 58% of the filters surveyed were changed out in the three year study period, and some 18% of all filters were changed out more than once. Most changeouts (63%) were due to the existence of a high pressure drop across the filter, indicative of filter plugging. Other reasons for change-out included leak-test failure (15%), preventive maintenance service life limit (13%), suspected damage (5%) and radiation buildup (4%). Filter failures occurred with approximately 12% of all installed filters. Of these failures, most (64%) occurred for unknown or unreported reasons. Handling or installation damage accounted for an additional 19% of reported failures. Media ruptures, filter-frame failures and seal failures each accounted for approximately 5 to 6% of the reported failures.

Based on the survey results, several general conclusions can be inferred, including:

1. HEPA filters have been generally performing the task they were designed for.
2. Most changeouts have been made because of filter plugging, preventive maintenance, or precautionary reasons rather than evidence of filter failure.
3. Where failures have been experienced, records generally have not been adequate to determine the cause of failure.
4. Where cause of failure has been determined, damage attributed to personnel handling and installation has been substantially more prevalent than that from filter environmental exposure. Some respondents indicated the need for improved personnel training in handling and installation.
5. Some reduction in filter failure frequency can be achieved by improving the acid and moisture resistance of filters, and providing adequate pretreatment of air prior to HEPA filtration.

Introduction

At the request of the Department of Energy (DOE), Airborne Waste Management Program Office and Exxon Nuclear Idaho Company, the Pacific Northwest Laboratory (PNL) performed a survey of high efficiency particulate filter applications and experience at DOE sites. While filter system design guidance has been available

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** Operated by Battelle Memorial Institute for the U.S. Department of Energy.

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for some years^(1,2), a literature survey⁽³⁾ revealed no substantial reports of filter changeout and failure experience. Therefore, the purpose of this survey was to provide an initial overview of the reasons and magnitude of HEPA filter changeout and failures as an aid in directing future HEPA filter technology development. Survey results were tabulated and presented in a PNL report.⁽⁴⁾ This paper summarizes some of the most significant findings.

Method

A questionnaire survey approach was selected as the method to gather HEPA filter experience data from a wide variety of DOE site applications. While extensive data were desired, it was recognized that a lengthy questionnaire would pose unreasonable demands upon recipients, and a low rate of return might result. Consequently, considerable effort was made to keep the questionnaire concise. The questionnaire developed was given a trial distribution within PNL and was found to be a reasonable request for information.

A total of 408 questionnaires were distributed via DOE Operations offices to 35 DOE site contractors reporting 1979 airborne releases to the DOE Effluent Information System data base. Data were requested for systems operated in the 1977-1979 period. Henceforth all discussion of results and all data tables are referenced to that three year period.

Usable data represented 24 DOE site contractors, 342 filter banks ranging from 1 to 790 filters each, and a total of 9154 filter applications within these banks. This data was entered into the PNL Biometrics Computer System and manipulated using the Minitab statistics package.⁽⁵⁾ Subjective comments received from contractor personnel were also noted.

Results

Twenty-six contractors returned 327 questionnaires for an approximate 80% return rate. Of the returned questionnaires, 223 (55% of those distributed) contained data suitable for use in the study. The remaining 104 questionnaires returned were excluded from the study due to lack of HEPA filters in the system, lack of operation in 1977-1979, or lack of data. Several site representatives indicated that compiling data to complete the questionnaires was a difficult process because records were not kept in a readily retrievable form or specific records were not available. One contractor indicated that no data regarding filter changeout or failure occurrences were available. In some cases, data were estimated by site representatives based on partial records and discussions with personnel directly involved in HEPA filter maintenance. Other sites did maintain records adequate for questionnaire completion.

The total numbers for HEPA filter applications, changeouts, and failures are listed in Table I. As used in this paper, an application is considered to be a single slot for a HEPA filter. A system consisting of one filter is considered a single application as is a single slot within a bank or array of filters.

The relatively low filter failure incidence (12%) over three years would appear to indicate that filters are generally performing their intended task. Furthermore, the ratio of total filters changed to filters failed (approximately 6 to 1) indicated that most filters are changed out prior to failure.

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Table I

HEPA Filter Experience Summary

<u>Description</u>	<u>Number</u>	<u>% of Total</u>
Filter Applications	9154	100%
Applications with no Changeouts	3870	42%
Applications in which Changeouts Occurred	5284	58%
Applications Having More than One Changeout	1610	18%
Filter Failures	1105	12%

Changeout Experience

Examining the incidence and reasons for filter changeouts gives further evidence that filters are performing well. Typical reasons for filter changeout include high differential pressure (ΔP) across the filter, failure to meet in-place leak test criteria, reaching a service life limit, suspected damage and radiation build up. This data is presented in Table II.

Table II

Filter Changeout Summary

<u>Changeout Reason</u>	<u>Number</u>	<u>% of Total</u>
High ΔP	4333	63%
Leak Test Failure	1020	15%
Preventive Maintenance Service Life	871	13%
Suspected Damage	376	5%
Radiation Buildup	256	4%
Other Reasons (Unspecified)	38	<1%
TOTAL	6894	100%

The largest majority (63%) of filter changeouts were attributed to high ΔP across the filter, indicative of filter plugging. This reason was four to five times higher than the next most prevalent reasons for changeout, which were leak-test failure and preventive maintenance service life limits. Suspected filter

damage due to facility mishaps, and build up of radiation levels or radioactivity (including criticality concerns) made relatively minor contributions to reasons for changeout.

Preventive maintenance service life changeouts are of special interest in Table II, because they assume that changeout is made only on the basis of filter age and not because of particular evidence of failure or plugging. The close approximation of service life changeouts to leak-test failure would seem to indicate that reasonably good judgement has been exercised in establishing preventive maintenance service lives.

Taken collectively, changeouts can be combined into two broad categories: Filters changed out to prevent anticipated failure; and filters changed out because they failed. (i.e., leaking filters.) From Table II it can be concluded that 85% of all changeouts were filters performing as intended, and the remaining 15% were not. This latter number compares favorably with the incidence of filter failure (12%) identified in Table I.

The tabulation of changeouts with filter stage is presented in Table III. The results were a little surprising since it was anticipated that first stages would be changed out substantially more often than final stages. However, when ratios of changeouts to filter applications are compared, such differences are not readily apparent. This could possibly be explained as a deficiency in the questionnaire which combined single stage and final stage filters together. In hindsight, combining single stage with first stage filters would have been a more logical choice, since neither circumstance would involve an upstream HEPA filtration stage.

Exposure of filters to a single significant environmental factor is tabulated with changeout incidence in Table IV. The vast majority of filters were reported as being exposed to no distinguishing environmental characteristics (i.e., they filtered essentially clean dry air environments as might be found in typical building ventilation systems or in systems with good pre-HEPA treatment features). The highest frequency of changeouts appeared to occur in hydrofluoric acid or high moisture environments. These environments also reflected the highest frequency of changeouts for leak-test failure and high ΔP . Improvement in the acid/moisture resistance of filters was the most frequently indicated development need, specifically cited by four sites. One site, a gaseous diffusion plant, identified specific problems with filtration of uranium hexafluoride (UF_6) hydrolysis products, including HF gas and particulate UO_2F_2 . The site reported laboratory studies showing slight decreases in filter efficiency after exposure to 3.2 grams of UF_6 with complete plugging after exposure to 34.7 grams. This site also indicated a study had shown that filters fell below 99.97% efficiency after exposure to 25 grams of HF (corresponding to hydrolysis of 100 grams UF_6) and to less than 68% efficiency after 41 grams HF exposure (equivalent to hydrolysis of 180 grams of UF_6).

Failure Experience

Filter failure modes and their reported incidence are listed in Table V.

TABLE III Stage Versus Changeouts (*)

Stage	Filter Applications	Total Changes	Leak Test	Other Penet.	High ΔP	Radiation Buildup	Suspect Damage	Service Life	Visual
All Changeouts	7205	6894 (0.96)	1020 (0.14)	15 (<0.01)	4333 (0.60)	256 (0.04)	376 (0.05)	871 (0.12)	23 (<0.01)
First of Series	1699	1803 (1.06)	210 (0.12)	--	1219 (0.72)	204 (0.12)	48 (0.03)	112 (0.07)	10 (0.01)
Intermediate	160	59 (0.37)	8 (0.05)	--	50 (0.31)	--	--	1 (0.01)	--
Single Stage or Final of Series	5346	5032 (0.94)	802 (0.15)	15 (<0.01)	3064 (0.60)	52 (0.04)	328 (0.05)	758 (0.12)	13 (<0.01)

(*) Entries are reported occurrences with calculated frequency ratio of changeout to corresponding filter applications given listed below each occurrence.

TABLE IV Single Environment versus Changeouts(*)

Environment	Filter Applications	Total Changes	Leak Test	Other Penet.	High ΔP	Radiation Buildup	Suspect Damage	Service Life	Visual
All Single Environments	4190	4042 (0.96)	623 (0.15)	14 (<0.01)	2059 (0.49)	192 (0.05)	376 (0.09)	766 (0.18)	12 (<0.01)
Solvent	5	4 (0.80)	--	--	4 (0.80)	--	--	--	--
High Moisture	82	327 (3.99)	60 (0.73)	--	134 (1.63)	107 (1.30)	1 (0.01)	25 (0.30)	--
High Dust	85	199 (2.34)	6 (0.07)	--	129 (1.52)	18 (0.21)	--	46 (0.54)	--
Grease/Oil	6	6 (1.00)	--	--	6 (1.00)	--	--	--	--
High Temperature	12	12 (1.00)	1 (0.08)	--	--	--	11 (0.92)	--	--
HF Acid	12	44 (3.67)	26 (2.17)	--	18 (1.50)	--	--	--	--
Other Acid	108	114 (1.06)	6 (0.06)	--	108 (1.00)	7 (0.06)	--	--	--
No Distinguishing Environment	3880	3336 (0.86)	524 (0.14)	14 (<0.01)	1660 (0.43)	67 (0.02)	364 (0.09)	695 (0.18)	12 (<0.01)

(*) Entries are reported occurrences with calculated frequency ratio of changeout to filter applications given below each occurrence.

Table V

<u>Filter Failure Modes</u>		
<u>Failure Mode</u>	<u>Number</u>	<u>% of Total</u>
Failure Mode Unknown	702	64%
Handling or Installation Damage	213	19%
Frame Failure	65	6%
Gasket or Seal Failure	62	6%
Media Rupture	54	5%
Filter Construction	6	<1%
Media to Frame Sealant Failure	<u>3</u>	<u><1%</u>
TOTAL	1105	100%

It is readily apparent from Table V that the large majority of failures occurred for unknown or unreported reasons. This would seem to indicate that most filter failures are either not investigated as to cause, or, if such an investigation is performed, it is not documented in a sufficiently retrievable manner.

Failure attributed to handling or installation damage accounted for 19% of the total failed filters. In addition to the raw data, three sites offered subjective comments indicating that improper filter handling and installation was a recurring problem, particularly with regard to inadequate or excessive clamping pressure. The use of wire mesh faceguards on filters was reported to have contributed to some reduction in handling damage. One site indicated a continued need for personnel trained in filter handling and testing. From these data and responses it would appear that improvements in personnel training might accomplish a significant reduction in filter failure frequency.

The incidences of frame failures, gasket or seal failures, and filter media ruptures were approximately equal; each constituted 5-6% of all filter failures. Where frame failure was identified, essentially all failed frames (58 out of 65) were wood. No observation of steel frame failure was reported, and the remaining 7 failures were of unspecified frame type. The predominance of wood frame failures could be due to frame warping or cracking caused by overtightening hold-down clamps.

Data concerning gasket or seal failure was somewhat inconclusive. Gasket seals were reported in much wider use (3920) than fluid seals (151), and proportionally more filters with gasket seals failed (422) than those with fluid seals (14). However, only 40 gasket seals and three (3) fluid seals were specifically identified as failing. Calculating the ratios of seal type failures to seal type applications can lead one to conclude that little difference may exist between gasket and fluid seal failure rates.

Media rupture could be due to a variety of causes including media breakdown, separator sagging, frame damage, or sealant failure. Possible differences in filter media were not addressed in this survey since it was assumed that essentially all filters used fiberglass paper. However, out of 54 reported occurrences of media rupture, several correlations with other filter component characteristics were noted. Specifically, all media ruptures occurred in wood-frame filters, and practically all (48) had aluminum separators and polyurethane foam sealants. Sealant failure was not identified as a significant filter failure mode (3 occurrences reported out of 1105).

A general lack of reported cases of filter failure attributed to faulty construction (6 in 1105) would appear to indicate that vendor and contractor quality control programs are collectively, functioning well.

Table VI presents failure modes as correlated with single characteristic environment exposure. Noteworthy in this table are the high frequencies of failure in hydrofluoric (HF) acid and high moisture environments. Ratios of filters failed to filter applications in which these failures were experienced are several times higher for HF acid and high moisture environments than for environments having no distinguishing characteristics, or the average of all single environment exposures. Also, the incidence of media rupture appears to occur most frequently in HF acid applications.

Service Life

Service life responses were received for 231 filter banks. Indefinite service life responses accounted for 53 of these banks. For the remaining 178 banks, a mean life of 3.0 years and standard deviation of 2.0 years was calculated. The distribution of these reported service lives is shown as a histogram in Figure 1.

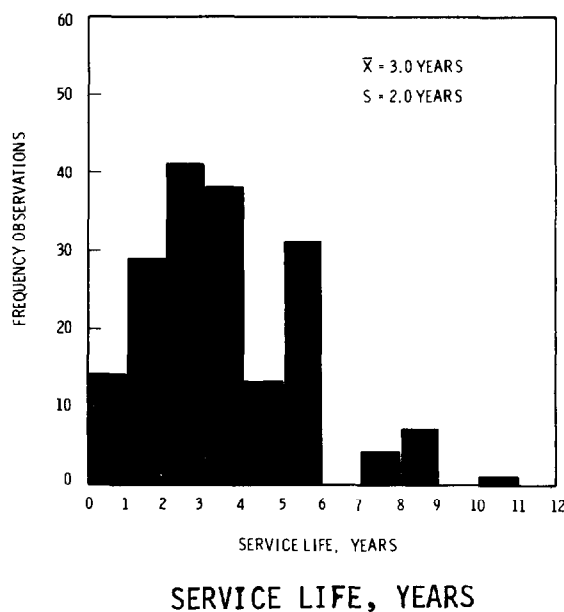


FIGURE 1. HEPA Filter Service Life

TABLE VI Single Environment versus Failure Mode (*)

Environment	Filter Applications	Filters Failed	Media Rupture	Sealant Failure	Frame Failure	Gasket or Seal Failure	Filter Construction	Handling or Installation Damage	Unknown
All Single Environments	2017	620 (0.31)	48 (0.02)	2 (0.001)	7 (0.003)	6 (0.003)	2 (0.001)	19 (0.009)	536 (0.27)
High Moisture	35	48 (1.37)	--	--	--	--	--	1 (0.029)	47 (1.34)
High Dust	1	1 (1.00)	--	--	--	1 (1.00)	--	--	--
High Temperature	6	1 (0.17)	--	--	--	--	--	--	1 (0.17)
HF Acid	12	26 (2.17)	26 (2.17)	--	--	--	--	--	--
Other Acid	63	12 (0.19)	--	--	--	--	--	10 (0.159)	2 (0.032)
No Distinguishing Characteristics	1900	532 (0.28)	22 (0.012)	2 (0.001)	7 (0.004)	5 (0.003)	2 (0.001)	8 (0.004)	486 (0.25)

(*) Entries are reported occurrences with calculated frequency ratio of failure mode to filter applications given below each occurrence.

Conclusions

The purpose of this survey was to provide an initial overview of the causes and frequencies of HEPA filter changeouts and failures. Since this was not intended to be a rigorous statistical study, caution must be exercised in the evaluation of the data presented. In this paper highlights of the original report⁽⁴⁾ have been presented. These highlights lead to the following general conclusions:

1. HEPA filters have generally been performing as designed.
2. Most changeouts have been made because of filter plugging, preventive maintenance and precautionary reasons rather than evidence of filter failure.
3. Where failures have been experienced, records generally have not been adequate to determine the cause of failure.
4. Where cause of failure has been determined, damage attributed to personnel handling and improper installation has been substantially more frequent than that from environmental exposure. Significant reduction in filter failure rate would be gained by improved worker training.
5. Some reduction in filter failure frequency can be achieved by improving the acid and moisture resistance of filters, and providing adequate pretreatment of air prior to HEPA filtration.

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DISCUSSION

FIRST: Are the changeout times local rules?

CARBAUGH: We did not specifically ask who defined the changeout times. Presumably, there are local rules.

MURROW: What were the capacities of the filters surveyed?

CARBAUGH: Flow was not a specific parameter requested in the questionnaire. Data for nuclear grade or application filters up to 24 in x 24 in. in size was requested.

ETTINGER: What was the range of typical service life change-out times?

CARBAUGH: Approximately three years with a standard deviation of two years.

HANSON: We changed from gasket type to fluid seal due to excessive frame leakage. In all, 280 filters were change in 1981.

CARBAUGH: Our survey was for 1977-1979, so your change was not reflected in our data.

DYMENT: Is it possible to derive filter plant operating efficiencies from the survey and thereby compare these with design efficiencies?

CARBAUGH: No. Specific efficiencies determined by filter tests were not requested in the survey.

BURCHSTED: What percent of the respondents test their filters routinely? How does this relate to the frequency of reported failures?

CARBAUGH: Frequency of testing was not requested so no correlation can be made with available data.

Appendix D

POTENTIAL APPLICATION OF A SINGLE PARTICLE AEROSOL SPECTROMETER
FOR MONITORING AEROSOL SIZE AT THE DOE FILTER TEST FACILITIES*

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Abstract

At each of the three Department of Energy (DOE) filter test facilities (FTF), a commercial single particle aerosol spectrometer, the LAS-X, was used to demonstrate that the test aerosol produced by each of the Q107 high efficiency particulate air (HEPA) filter test systems was polydisperse with a geometric standard deviation greater than 1.35, and a geometric mean diameter less than 0.2 μm . These results confirm the work of others that the "OWL" (mechanical analyzer) is inadequate for assuring that these test aerosols are monodisperse with a mean diameter of 0.3 μm . Use of the LAS-X for monitoring aerosol size was evaluated. The LAS-X is relatively insensitive to changes in aerosol concentration and refractive index over the range of interest for filter testing. It appears that the LAS-X would provide a better instrument for monitoring aerosol size, and would permit the FTF test operator to adjust the test aerosol to approximate the size characteristics currently recommended for this test.

I. Introduction

HEPA filters are routinely quality assurance (QA) tested at one of the three DOE FTF. This QA test includes evaluation of filtration efficiency against a monodisperse aerosol assumed to be 0.3 μm diameter, to assure that a minimum filtration efficiency requirement of 99.97 per cent for this aerosol is satisfied. It has been frequently assumed that 0.3 μm represents the size of maximum penetration, and these tests represent a conservative evaluation of actual filter performance. Actually the size of maximum penetration may be less than 0.3 μm . Size of the test aerosol is determined by measurement of the polarization ratio of the light scattered at right angles to an incident light beam using a device called the "OWL" or mechanical analyzer. The theoretical basis for this device, and experimental studies indicate that accurate size characterization of polydisperse aerosols cannot be made with the OWL(1,2). Because of this limitation to using the OWL, and concern that the QA test aerosols may not be monodisperse, a test program was performed to (1) determine the size characteristic of the FTF QA test aerosol and (2) evaluate the possible application of a single particle aerosol spectrometer (LAS-X) to replace the OWL. If the LAS-X is usable, it might permit the FTF operator to better control the size of the test aerosol.

*Work supported by the Department of Energy, Nuclear Fuel Cycle and Waste Management Division.

II. Single Particle Aerosol Spectrometer

Aerosol size distribution can be completely characterized by the geometric mean diameter or count median diameter (d_g) and geometric standard deviation (σ_g) if the aerosol has a log-normal size distribution. One way to determine these size distribution parameters is to use a single particle aerosol spectrometer to measure the light scattered by individual particles as they pass through a laser beam and use these data to generate a frequency histogram. In a single particle aerosol spectrometer, such as the one shown in Fig. 1, the sample stream containing the aerosol is constrained to a narrow cylinder in the center of a particle free sheath air stream. The concentric streams then pass through a narrow orifice where the aerosol particles accelerate and then pass in single file through the center of a laser beam. In the spectrometer shown in Fig. 1, part of the light scattered by an aerosol particle is collected by a mirror lens system and focused onto a photodetector. Over the range of interest ($0.09 \mu\text{m} - 0.8 \mu\text{m}$), the photodetector signal is a monotonic function of the diameter of the particle (see Fig. 2). The peak signal is quantized and added to one bin of a pulse height analyzer. The bins are arranged in order of increasing diameter. The pulse height analyzer builds up a frequency histogram giving the size distribution of the aerosol. These data can be transferred to a desktop computer where the geometric mean diameter and geometric standard deviation are computed and the histogram displayed and plotted immediately after the data are acquired.

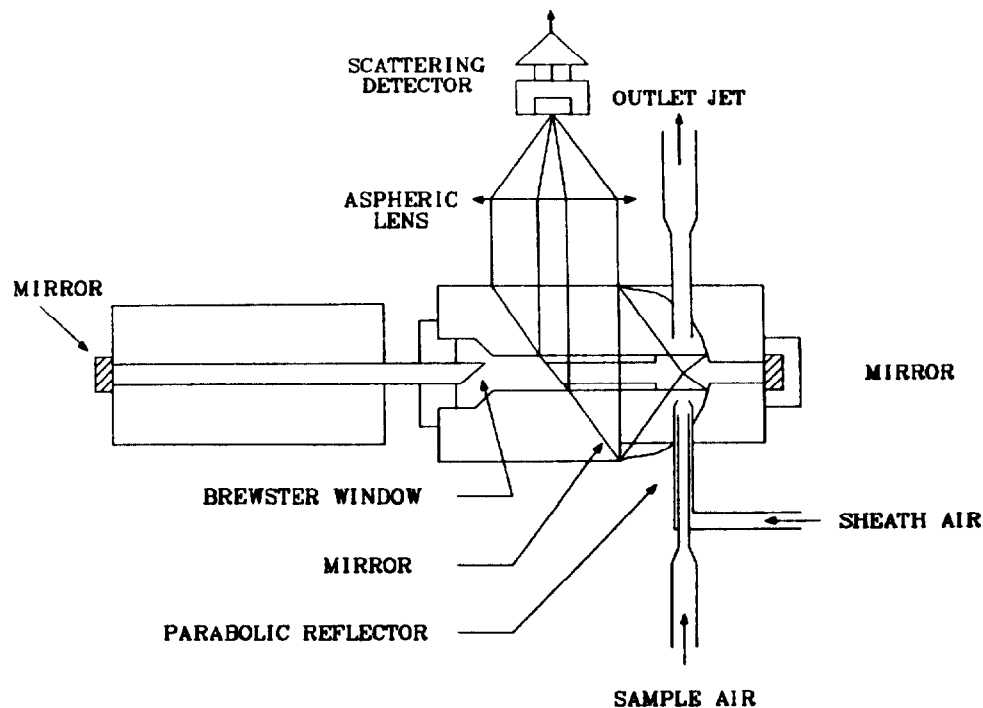


FIGURE 1
SCHEMATIC DIAGRAM OF THE OPTICAL SYSTEM FOR THE LAS-X SINGLE PARTICLE AEROSOL SPECTROMETER SHOWING THE INTRACAVITY EXCITATION AND AERODYNAMICALLY FOCUSED SAMPLE INLET.

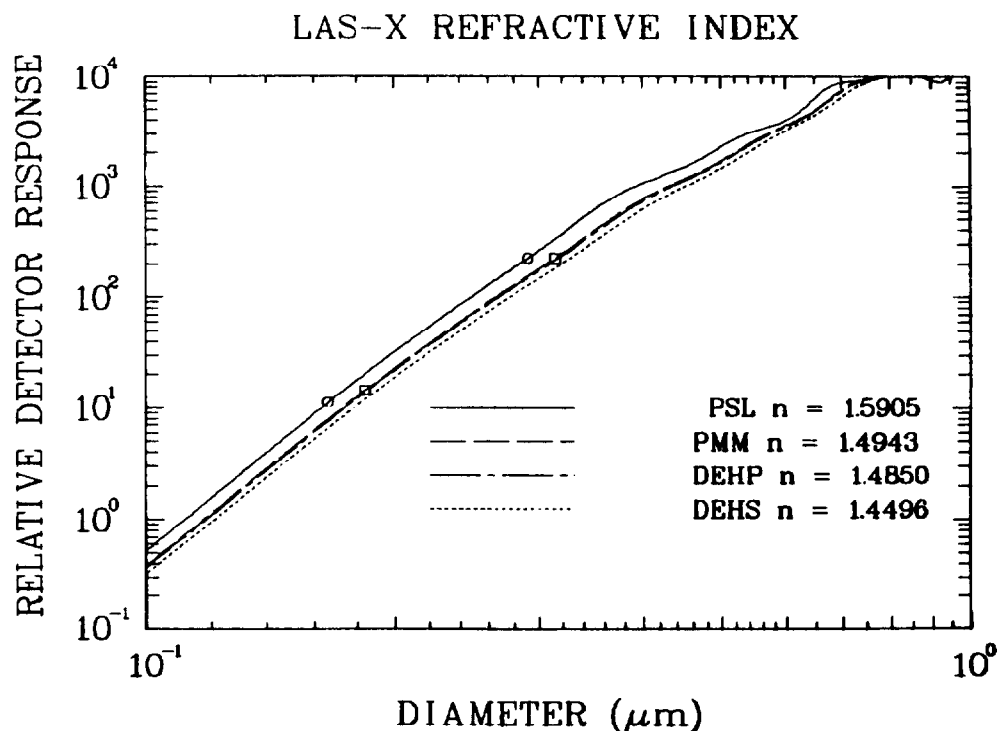


FIGURE 2

RELATIVE LAS-X DETECTOR RESPONSE VERSUS PARTICLE DIAMETER (CALCULATED) FOR DIFFERENT PARTICLE REFRACTIVE INDICES. PSL IS POLYSTYRENE LATEX. THE TWO CIRCLES ARE EXPERIMENTAL VALUES FOR 0.176 AND 0.312 μm DIAMETER PSL PARTICLES. PMM IS POLYMETHYL METHACRYLATE. THE TWO SQUARES ARE EXPERIMENTAL VALUES FOR 0.17 AND 0.325 μm DIAMETER PMM PARTICLES. DEHS IS DIETHYLHEXYL SEBECATE. THE CURVE FOR DEHP IS INDISTINGUISHABLE FROM THAT FOR PMM.

The principal advantage of the LAS-X over other optical single particle spectrometers is the fact that the particles are detected inside the laser cavity. This provides a factor of at least 6 increase in laser intensity over external illumination and standing wave illumination (from both sides) which significantly reduces the sensitivity of the LAS-X to particle refractive index (3). The LAS-X is advertised as being useful over the size range of 0.09 to 3.0 μm . Since the range of interest for QA testing at the FTF is 0.1 to 0.8 μm , this instrument appeared to provide a potentially useful tool for use at the FTF. While the LAS-X has been described in the literature (3,4,5), it was considered necessary to evaluate its operational characteristics at the FTF. Of special interest were the effects of (a) index of refraction, and (b) aerosol concentration.

III. Mechanical Analyzer (OWL)

Size of the FTF QA test aerosol is determined by measurement with a mechanical analyzer (OWL) which operates on the principle that the light scattered by a transparent, colorless, spherical aerosol particle illuminated from one direction with unpolarized light is partially polarized (see Fig. 3). The scattered light consists of two plane polarized components with their planes of polarization at right

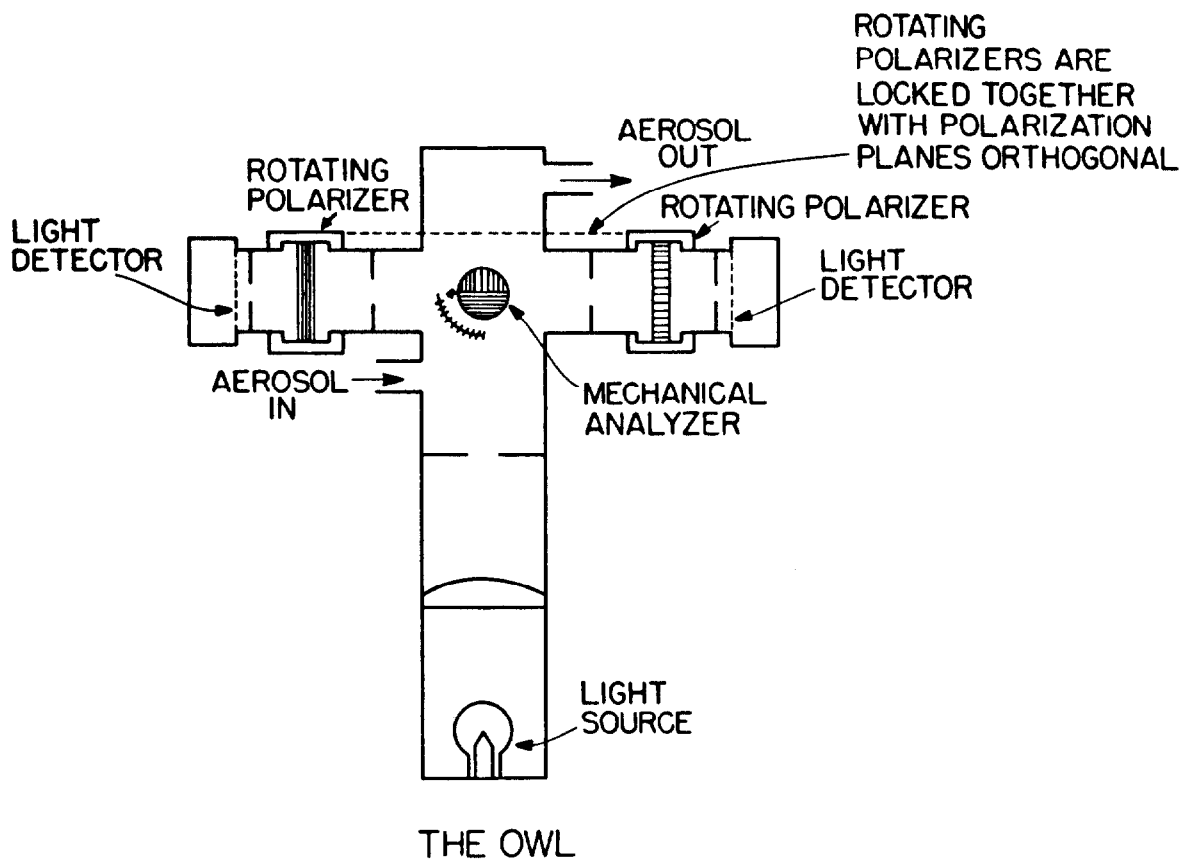


FIGURE 3

SCHEMATIC DIAGRAM OF THE OWL (MECHANICAL ANALYZER) SHOWING THE SOURCE OF UNPOLARIZED LIGHT AT THE BOTTOM AND THE "VISUAL OWL" MECHANICAL ANALYZER VIEWING THE LIGHT SCATTERED FROM THE AEROSOL AT 90°. IN THE PHOTOELECTRIC VERSION OF THE OWL THE ROTATING POLARIZERS ON THE ARMS OF THE T ARE LOCKED TOGETHER WITH THEIR POLAROID AXES ORTHOGONAL.

angles to each other. The mechanical analyzer measures the ratio of these two components at 90 degrees to the incident beam of light to determine the particle size. The polarization ratio is sensitive to refractive index and is a steep function of particle diameter. Theoretical curves are available to relate this measurement to particle size for a particle of given index of refraction.

The scattering plane is defined by the direction of the incident beam and the direction of the scattered light ray to a detector. Scattered light polarized perpendicular to this plane is designated by intensity i_1 , and light polarized parallel to this plane is designated as intensity i_2 . The mechanical analyzer contains two photodetectors in the same plane at 90° to the incident light beam. This orientation results in the photodetectors being 180° apart. The polarizers in front of each detector are mechanically locked together so that their polarization axes are perpendicular. One detector measures the total i_2 intensity and the other measures the total i_1 intensity. The analyzer angle, θ , is given by: $i_2/i_1 = \tan^2 \theta$. For a monodisperse diethylhexyl phthalate (DEHP) aerosol of refractive index 1.485 and median diameter 0.3 μm , the mechanical

analyzer will exhibit a null reading at an analyzer setting of 29-30 degrees where the light intensity penetrating each polarizer is equal. As for the LAS-X, instrument response is dependent on the index of refraction of the aerosol material.

The limitations of the OWL when used to characterize the size of an aerosol that is not monodisperse is indicated by the following example. A mixture of 80 per cent 0.15 μm diameter DEHP, and 20 per cent 0.30 μm diameter DEHP, would exhibit a null reading at an analyzer setting of 28 degrees. If one assumed this was a monodisperse aerosol, these data would be interpreted to show a diameter slightly larger than 0.29 μm . This false indication of aerosol size is due to the large particles scattering significantly more light at 90 degrees than do small particles. Thus, the smaller particles contribute only a small fraction of the detected intensities i_1 and i_2 , and as a result the mechanical analyzer gives erroneous readings when examining polydisperse aerosols. As discussed later in this report, polydisperse aerosols are generated at the FTF. For the mechanical analyzer to provide meaningful results, the aerosol being sized must be monodisperse. In their report describing the OWL, LaMer and Sinclair⁽⁶⁾ emphasized that the aerosol must be "moderately homogeneous as to particle size" for the OWL to give a correct size measurement. Using a thermal (Q-127) DEHP aerosol generator, Hinds, et al⁽¹⁾ showed that the OWL gives an incorrect measurement of the aerosol geometric mean diameter when the aerosol is polydisperse. Gerber⁽²⁾ showed that for an OWL reading of 29° the geometric mean diameter is a rapidly decreasing function of DEHP geometric standard deviation. In particular the OWL reads 29° for a DEHP aerosol with geometric mean diameter 0.23 μm and geometric standard deviation of 1.3. As indicated later in this report the Q-107 aerosol geometric standard deviations observed at the three DOE filter test facilities are all in excess of 1.3.

IV. Experimental Approach

Evaluation of the LAS-X

Since the LAS-X is a single particle detector, the simultaneous presence of two particles in the sensitive volume will result in erroneous data. To handle the concentrations encountered at the test systems, a variable aerosol diluter which can operate in the range of 200:1 up to 1000:1 was constructed for use with the LAS-X at each FTF⁽⁷⁾. With the diluter operated at a dilution ratio between 250:1 and 1000:1, the LAS-X can sample the aerosol size distributions at the 100 microgram per liter concentrations specified for the FTF test systems without interference due to coincidence. Under these test conditions, the aerosol concentration is less than 1500 particles/channel/sec or 20000 total particle/sec. This is within the limits advertised by the manufacturer (9000 particles/channel/sec or 25000 total particles/sec). Even so, we will later discuss our verification of use of the LAS-X at these concentrations. The LAS-X is calibrated with a series of highly monodisperse polystyrene latex microspheres⁽⁴⁾ covering the diameter range 0.1 μm to 3.0 μm . The signals are then least squares fit using Mie Theory⁽⁸⁾ to enable interpolation between the calibration points. The voltage bin widths in the LAS-X pulse height analyzer are then set with precision resistors so that

within each of the four size ranges ($0.09\text{ }\mu\text{m} - 0.195\text{ }\mu\text{m}$, $0.15\text{ }\mu\text{m} - 0.3\text{ }\mu\text{m}$, $0.24\text{ }\mu\text{m} - 0.84\text{ }\mu\text{m}$, $0.60\text{ }\mu\text{m} - 3.0\text{ }\mu\text{m}$) each of the 15 size bins measure equal diameter intervals. This calibration can be checked readily using a set of Dow polystyrene latex calibration spheres⁽⁹⁾. The mean diameters (μm) available at each FTF are as follows: 0.109, 0.220, 0.312, 0.497, and $1.091\text{ }\mu\text{m}$.

Light scattering measurements are, in general, sensitive to the refractive index of the scattering object. Fig. 2 shows the calculated LAS-X scattered light detector response as a function of spherical particle diameter for three different particle indices of refraction corresponding to polystyrene latex microspheres ($n = 1.5905$), polymethyl methacrylate ($n = 1.4943$), diethylhexyl phthalate (DEHP) ($n = 1.485$), and diethylhexyl sebecate (DEHS) ($n = 1.4496$) which is being used as a replacement for DEHP at one FTF because of a concern over the potential carcinogenicity of DEHP.

Different scattering intensities are produced by particles of the same size having different refractive indices. While the instrument is calibrated in terms of particle size using polystyrene latex microspheres, a simple algorithm based on Mie Theory (Fig. 2) can be used to permit direct calculation of the diameters of the test aerosols for the aerosol size of concern even though the calibration aerosol has a different index of refraction. Mie Theory represents a rigorous solution to the light scattering problem for spherical droplets or particles under conditions of plane wave illumination. Although these conditions are not met exactly in the LAS-X, the approximation is very good. Other approximations include Rayleigh scattering and Rayleigh-Gans-Debye scattering⁽⁸⁾. For a wave length of $0.633\text{ }\mu\text{m}$, Rayleigh theory is applicable only below a diameter of $0.06\text{ }\mu\text{m}$. Rayleigh-Gans-Debye theory is applicable only for diameters much less than $0.2\text{ }\mu\text{m}$ for DEHP and is, therefore, inappropriate for any calculation involving the LAS-X.

An additional series of tests using aerosols of known size and a different index of refraction was performed using polymethyl methacrylate (PMM), index of refraction 1.4943, to confirm the calculated extrapolation from the PSL curve. Figure 2 shows the calculated curve for PMM together with the data points for two PMM aerosols. In the region of $0.3\text{ }\mu\text{m}$ diameter PSL spheres of $0.312\text{ }\mu\text{m}$ nominal diameter and PMM spheres of nominal diameter $0.325\text{ }\mu\text{m}$ both produced LAS-X modal diameter values of $0.30\text{ }\mu\text{m}$. The corrected PMM sphere diameter was $0.313\text{ }\mu\text{m}$. PSL spheres of nominal diameter $0.176\text{ }\mu\text{m}$ gave a LAS-X modal diameter of $0.155\text{ }\mu\text{m}$ while PMM spheres of nominal diameter $0.17\text{ }\mu\text{m}$ produced a LAS-X modal diameter of $0.16\text{ }\mu\text{m}$. When this point was extrapolated to the PMM curve using the same normalization as required to place the $0.155\text{ }\mu\text{m}$ diameter PSL particle on its Mie Theory curve, the signal value differed from the Mie Theory estimate by only 1.25 per cent and the corrected diameter was $0.169\text{ }\mu\text{m}$. Agreement between the data points and the calculations substantiate the use of Mie Theory to extrapolate LAS-X data to aerosols with different refractive indexes. Figure 4 shows the LAS-X histogram for nominal $0.312\text{ }\mu\text{m}$ diameter PSL particles. Figure 5 shows that for nominal $0.176\text{ }\mu\text{m}$ diameter PSL particles. In these figures the solid curve is for range 2 and the dashed curve is for range 3 of the LAS-X.

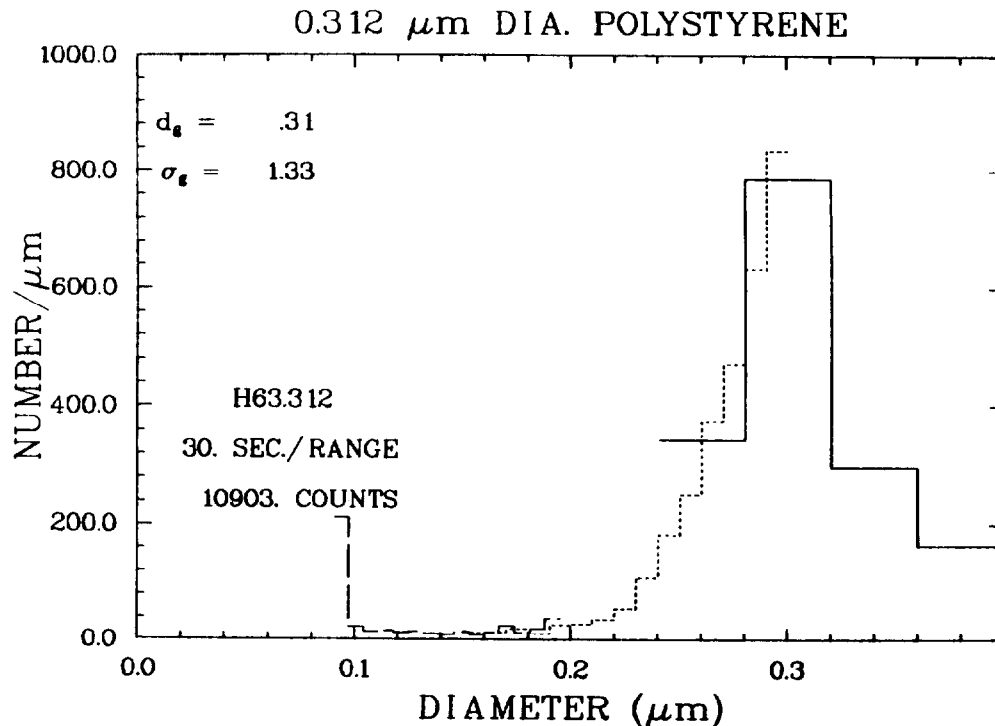


FIGURE 4

HISTOGRAM SHOWING PARTICLE FREQUENCY AS A FUNCTION OF PARTICLE DIAMETER FOR AN AEROSOL OF 0.312 μm DIAMETER (NOMINAL) POLYSTYRENE LATEX SPHERES. ONLY THE TWO SMALLEST SIZE RANGES ARE SHOWN BECAUSE THERE WERE VERY FEW PARTICLES LARGER THAN 0.4 μm DIAMETER. SCANNING ELECTRON MICROSCOPY (SEM) GAVE A GEOMETRIC MEAN DIAMETER OF 0.28 μm AND A GEOMETRIC STANDARD DEVIATION OF 1.22.

Concentration Effects

For a single particle aerosol spectrometer like the LAS-X to operate properly, there must be only one particle at a time in the laser beam and the processing electronics must recover to a baseline value between particles. The manufacturer claims that the maximum count rate is limited by the baseline restorer electronics to the lesser of 9000 counts/sec/channel or 25000 total counts/sec over the 16 channels in the pulse height analyzer. For this reason an aerosol diluter must be used with the LAS-X at the FTF. The effect of aerosol concentration on the LAS-X response was evaluated using a DEHS aerosol at various concentrations ranging from 0.04 mg/m^3 to 0.18 mg/m^3 . At a dilution of 500:1 this represents an aerosol concentration of 20-90 mg/m^3 . A series of three independent tests involving 9 different concentrations for each test showed a coefficient of variation in measured geometric mean diameter of less than 1 per cent. Fig. 6 is a plot of geometric mean diameter of the DEHS aerosol as a function of dilution ratio, which is the aerosol concentration at the diluter inlet divided by that at the diluter outlet. Note that the geometric mean diameter is beginning to increase at the low dilution ratio end of the scale. At a 200:1 dilution ratio the total count rate is 4300 particles/sec.

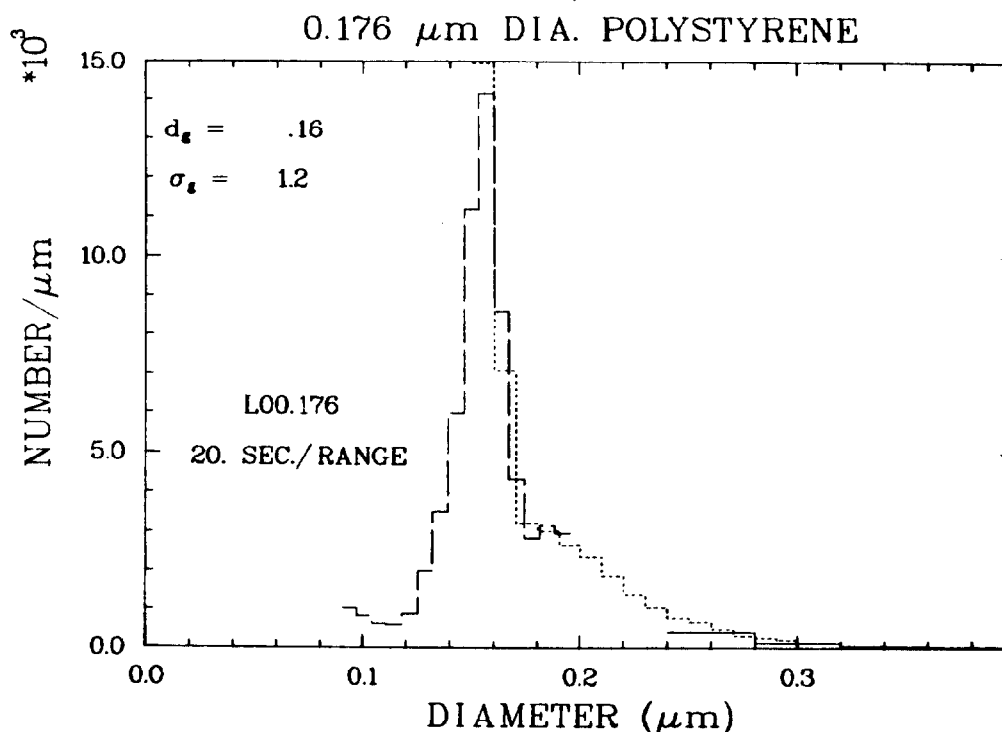


FIGURE 5

HISTOGRAM SHOWING PARTICLE FREQUENCY AS A FUNCTION OF PARTICLE DIAMETER FOR AN AEROSOL OF 0.176 μm DIAMETER (NOMINAL) POLYSTYRENE LATEX SPHERES. ONLY THE TWO SMALLEST SIZE RANGES ARE SHOWN BECAUSE THERE WERE VERY FEW PARTICLES LARGER THAN 0.4 μm DIAMETER. SEM ANALYSIS GAVE A GEOMETRIC MEAN DIAMETER OF 0.18 AND A GEOMETRIC STANDARD DEVIATION OF 1.09.

Aerosol Characterization at FTF

Test aerosol size distribution data were obtained at each FTF during routine QA testing of 1000 and 575 CFM HEPA filters. Calibrated aerosol diluters and LAS-X instruments were installed at each FTF. In each case the Q107 was adjusted by the operator to deliver smoke of 0.3 μm geometric mean diameter according to the mechanical analyzer (OWL). Figs. 7, 8, and 9 show typical aerosol size distributions selected from measurements over several days at the three FTF. The background count rate was less than 9 counts/sec summed over all channels. These data indicate that the aerosol geometric mean diameter is smaller than 0.2 μm , with a σ_g ranging from 1.35 to 1.45, well in excess of the generally accepted definition for a monodisperse aerosol.(10)

At these σ_g values, the OWL cannot uniquely define the size of the test aerosol. At a σ_g value of 1.3 an OWL reading of 29° is produced by an aerosol with a geometric mean diameter of 0.23 μm (2).

These results at the FTF, and other informally reported data evaluating the monodispersity of thermally generated aerosols in systems of this type suggest that recently developed instrumentation such as the LAS-X provides a more accurate means for measuring aerosol size

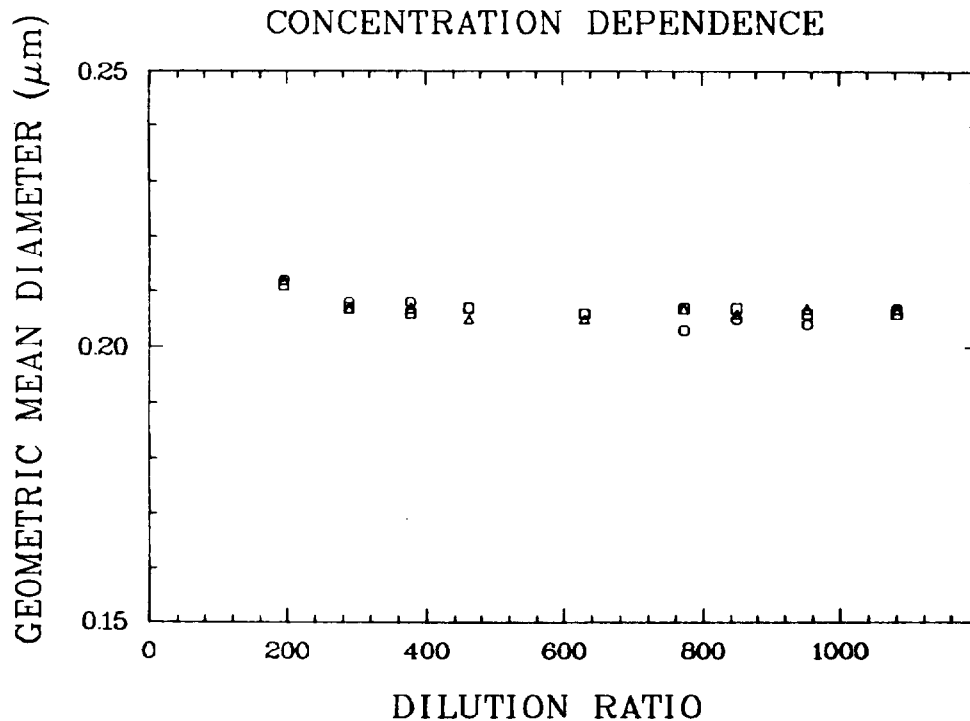


FIGURE 6

CONCENTRATION DEPENDENCE OF THE GEOMETRIC MEAN DIAMETER OF A DEHS AEROSOL AS DETERMINED BY THE LAS-X. A DILUTION RATIO OF 1000:1 CORRESPONDS TO 0.04 mg/m^3 . AT 200:1 THE TOTAL COUNT RATE WAS 4300 PARTICLES/SEC. THE SQUARES, CIRCLES AND TRIANGLES REPRESENT THREE INDEPENDENT EXPERIMENTAL RUNS.

characteristics, and also permitting the FTF operator to better control aerosol size.

V. Discussion

The existing FTF QA test aerosols measured during this study do not meet the DOE test specifications in that a) the test aerosols are not monodisperse based on the usually accepted definition for a monodisperse aerosol(10) and b) the test aerosols do not have a $0.3 \mu\text{m}$ geometric mean diameter (geometric mean diameter was less than $0.2 \mu\text{m}$).

The mechanical analyzer (OWL) now used to monitor aerosol size cannot accurately characterize a polydisperse QA test aerosol. Therefore, the FTF test personnel cannot adjust the aerosol to meet the existing aerosol test specifications. While review of the military specifications(11) indicates that the test aerosol is only specified in terms of an OWL reading of $29-30^\circ$, the generally accepted definition of the intention of this specification is to provide a $0.3 \mu\text{m}$ monodisperse test aerosol.

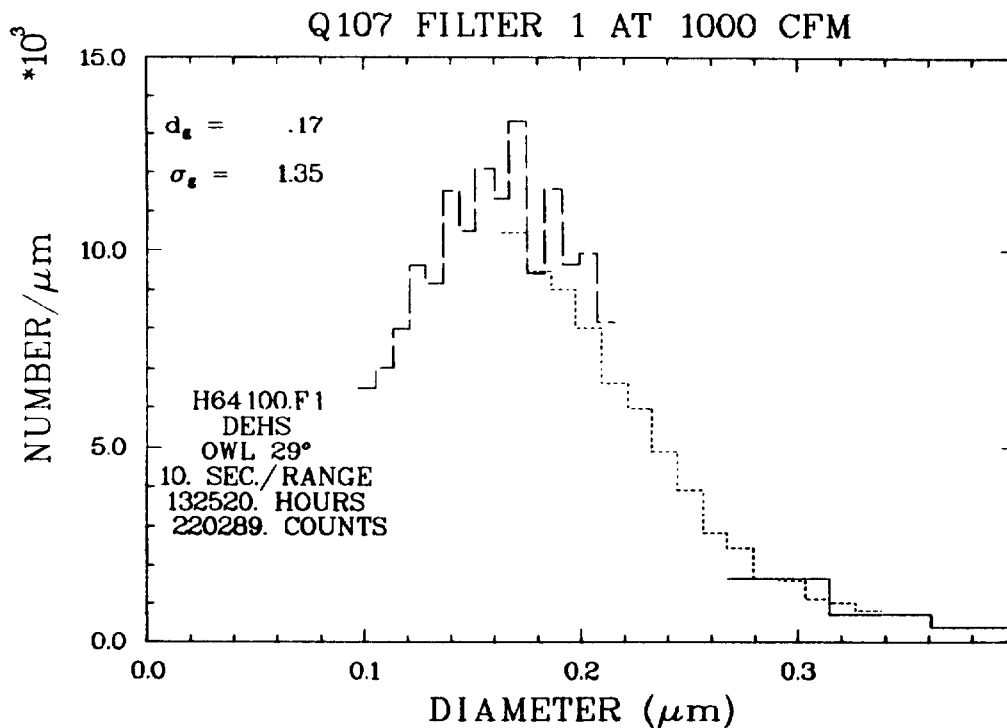


FIGURE 7

SIZE DISTRIBUTION OF DEHS AEROSOL MEASURED AT THE HANFORD FTF. THE THREE LINE TYPES REPRESENT RANGES 1, 2, AND 3 FOR THE LAS-X. THE BAR WIDTHS REPRESENT THE RESPECTIVE HISTOGRAM BIN WIDTHS FOR EACH RANGE. THE GEOMETRIC MEAN HAS BEEN CORRECTED FOR THE REFRACTIVE INDEX DIFFERENCE BETWEEN PSL AND DEHS.

Accurate information regarding the size of the test aerosol can be obtained with the use of commercially available instrumentation such as the LAS-X to monitor aerosol geometric mean diameter and geometric standard deviation. The LAS-X may be a particularly good choice because of its intracavity sensing, which provides high laser intensity and minimizes corrections for refractive index effects. Experimental tests indicate that the index of refraction correction can be applied and concentration effects are not significant with the use of a simple dilution system. For the proposed application, this diluter does not have to be accurate since we are not trying to quantify aerosol concentration. The diluter must only reduce the aerosol concentration below the level where coincidence loss is significant.

As it is now configured the LAS-X data output is a paper tape record containing numerical values which can be plotted to give a histogram of the aerosol size distribution, assuming the aerosol has the same refractive index as that of the polystyrene latex calibration particles. Used in this manner, the LAS-X would not provide the FTF with a real time monitor indicating geometric mean diameter and geometric standard deviation. This situation has been remedied by adapting a relatively inexpensive state-of-the-art desktop microcomputer to the LAS-X so that the FTF operators will have a real time display of the aerosol size distribution, corrected for refractive index. Test

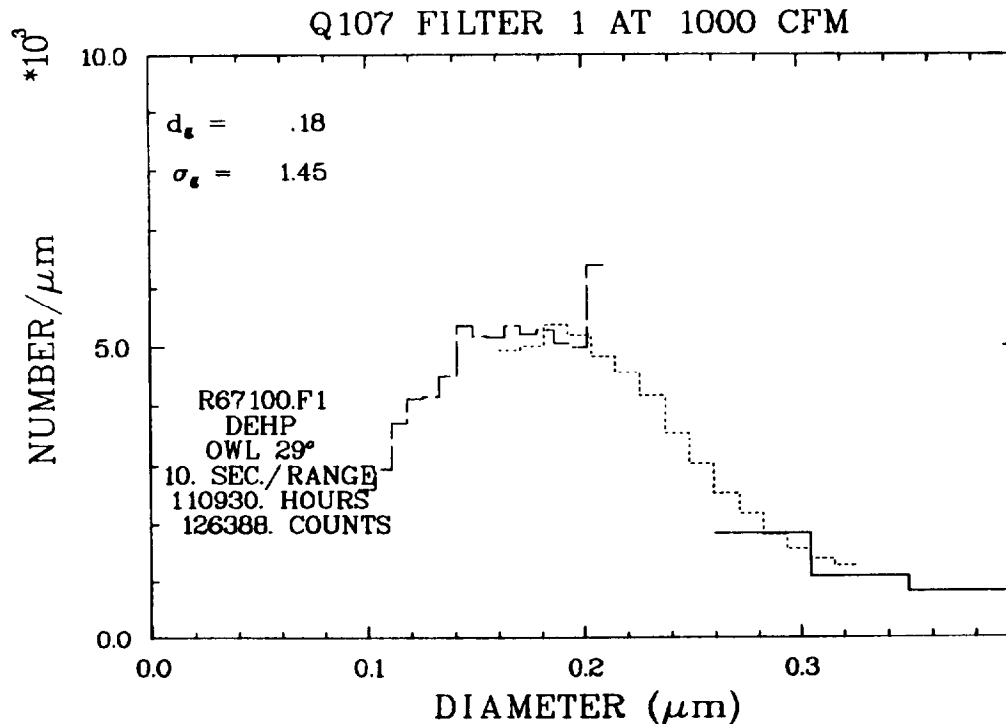


FIGURE 8

SIZE DISTRIBUTION OF DEHP AEROSOL MEASURED AT ROCKY FLATS FTF. THE THREE LINE TYPES REPRESENT RANGES 1, 2, AND 3 OF THE LAS-X. THE BAR WIDTHS REPRESENT THE RESPECTIVE HISTOGRAM BIN WIDTHS FOR EACH RANGE. THE GEOMETRIC MEAN HAS BEEN CORRECTED FOR THE REFRACTIVE INDEX DIFFERENCE BETWEEN PSL AND DEHP.

aerosol geometric mean diameter and geometric standard deviation are directly indicated. The desktop computer evaluated at Los Alamos is the Hewlett Packard HP-85 which has a small high resolution CRT display, a dot-matrix thermal printer which can copy images from the CRT display and a tape cassette on which to store data. The cost of this device is approximately \$3500. It weighs only 20 pounds.

This package will permit each FTF, regardless of the specific test aerosol used at each station, (e.g. DEHP or DEHS) to adjust their test systems in an attempt to better approximate an $0.3 \mu\text{m}$ monodisperse test aerosol. It may still be difficult to consistently attain this goal with the existing aerosol generation system due to the difficulties of generating a monodisperse aerosol over long time periods using a relatively large test system such as the Q107.

The LAS-X instrument has yet to be evaluated for its ruggedness in field use. The LAS-X at Los Alamos has experienced both power supply and laser tube failure during the past year. This evaluation should be performed during parallel operation of the OWL and LAS-X at one or more FTF.

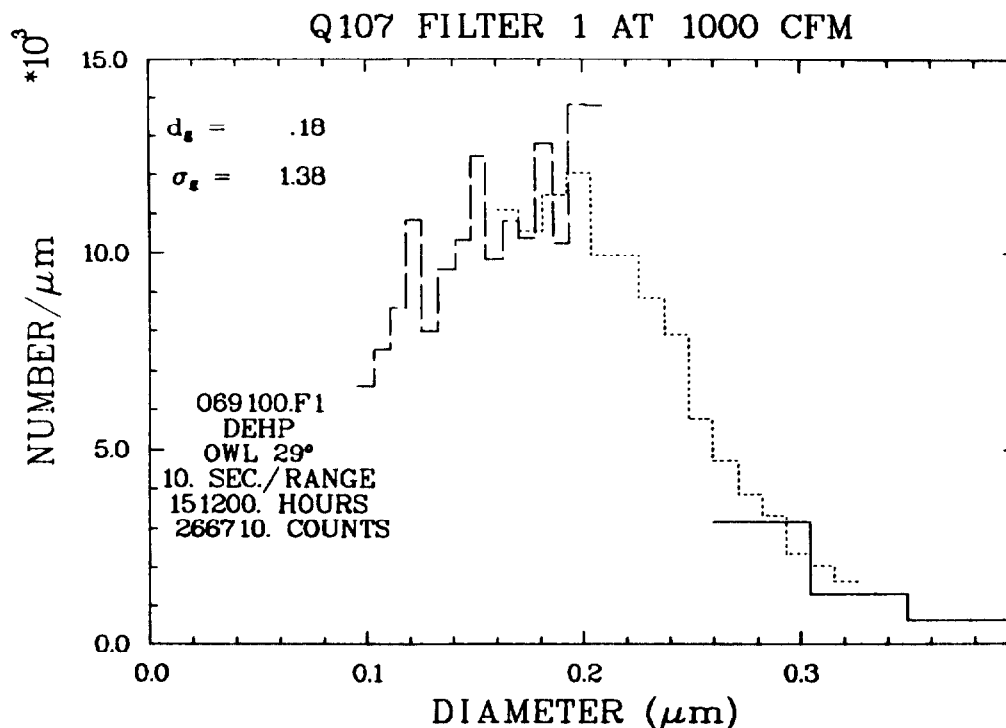


FIGURE 9

SIZE DISTRIBUTION OF DEHP AEROSOL MEASURED AT OAK RIDGE FTF. THE THREE LINE TYPES REPRESENT RANGES 1, 2, AND 3 OF THE LAS-X. THE BAR WIDTHS REPRESENT THE RESPECTIVE HISTOGRAM BIN WIDTHS FOR EACH RANGE. THE GEOMETRIC MEAN HAS BEEN CORRECTED FOR THE REFRACTIVE INDEX DIFFERENCE BETWEEN PSL AND DEHP.

Summary

A single particle aerosol spectrometer, the LAS-X has been evaluated for use in measuring the size distribution of the test aerosol in the Q107 aerosol generators at the three DOE FTF. The LAS-X data indicate that the aerosol at the FTFs is polydisperse with a geometric mean diameter less than $0.2 \mu\text{m}$ and a geometric standard deviation in excess of 1.3. This observation calls into question the use of the OWL to monitor the test aerosol size. The refractive index dependence of the LAS-X was shown to be small and to be easily correctable. The concentration dependence was shown to be minimal over the normal LAS-X operating range. A microcomputer was adapted to the LAS-X to give a real time display of the aerosol size distribution and a display of the geometric standard deviation and geometric mean diameter. A single particle aerosol spectrometer such as the LAS-X can be incorporated readily into the test system as a replacement for the OWL for measurement of the test aerosol size distributions.

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DISCUSSION

FIRST: In looking at the curves which you demonstrated from each of the three test stations, it seemed to me that the particle sizes were discontinuous and arranged into rather discrete peaks. Can you explain why this occurs? One usually assumes there would be smooth curves because of the process of formation. Is this an artifact of the instrument, or do you believe that this is an accurate representation of the size?

SALZMAN: I think a good smooth log-normal distribution is a more accurate representation of the size. One reason that discontinuities can occur is if the bin widths were not exactly the same. In other words, across range three, where you see this problem most acutely, the bin width is supposed to be $0.007 \mu\text{m}$. If there was a variance, say one of the bins is poking up higher plus a little wider, (and, of course, I have no easy way to check the bin widths) I just have to assume that $0.007 \mu\text{m}$ is correct and adjust my data accordingly. That accounts for the variation. I think I have enough counts so that it isn't poor counting statistics that account for the peaks. It is probably slight variations in bin widths that give rise to that sort of thing. I don't believe that any of the peaks represent real data.

MURROW: Now that it has been determined that most filters are being tested with $0.17 \mu\text{m}$ aerosols, have you or others determined what size particle is most penetrating?

SALZMAN: No, but we could do this as part of our collaborative study with investigators at The Inhalation Toxicology Research Institute in Albuquerque.

ETTINGER: In reply to the question by Jack Morrow, we at LANL have done no recent work to define the size of maximum filter penetration. However, the IAS-X could be used for this purpose.

GERBER: Did the estimates of the mean and geometric standard deviation take into account the truncation of the distribution at smaller sizes (those less than $0.09 \mu\text{m}$)?

SALTZMAN: The cutoff at $0.09 \mu\text{m}$ results in less than 4% overestimate of geometric mean diameter and 4% underestimate of geometric standard deviation.

BERGMAN: The dilution system required for the laser particle analyzer has a significant effect on the particle size distribution, i.e., diluting aerosols will result in a disproportionate loss of large particle sizes. For $5.0 \mu\text{m}$ particles and a dilution ratio of 500, we have a particle loss of 99%. It is around 10% for $1 \mu\text{m}$ particles.

TILLERY: Particle losses in the diluter can become significant if particle size is large. The maximum size for the measured distribution is about $0.4 \mu\text{m}$. In this size range the losses would not be significant. Aerosols containing particles much above $1.0 \mu\text{m}$ in

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diameter would require corrections for losses.

FIRST: But, Dr. Salzman was saying that the preferred method is to use a polydisperse aerosol, I think that your argument does not hold for that type of aerosol.

TILLERY: He asked what effect the diluter had on the aerosol we measured. You can see that the aerosol size tails off above 0.3 μm . That is the aerosol I am talking about.

FIRST: Perhaps you can comment on my question.

TILLERY: If you can use a polydisperse aerosol that has a very wide size distribution, you would have to calibrate the diluter with respect to losses for large particles. I certainly agree with that.

FIRST: What would be your standard error? Would you care to estimate it?

TILLERY: I would have to know what size distribution aerosol you are talking about.

FIRST: Cold DOP.

TILLERY: Cold DOP is 0.6 μm on median size with a geometric standard deviation about 1.4.

FIRST: It is probably more like 1.65.

SALTZMAN: The losses of the large particles would be less than 15%.

FIRST: Isn't that a fairly significant loss?

SALTZMAN: Certainly it is a significant loss. But you have to calibrate the unit and make appropriate corrections.

STEINBERG: Is the laser to replace the mechanical analyzer at a cost of \$17,000 vs. \$4,000?

SALTZMAN: Since the mechanical analyzer does not work for polydisperse aerosols, it should not continue to be used to control the aerosol size at the Filter Test Facilities. A large number of different size distributions will give the same mechanical analyser reading. The cost difference is not significant when considered in light of the cost of a Q107 which is supposed to generate a monodisperse aerosol.

STEINBERG: Has anyone thought of the significance of the results that you are getting? Over the years we have been led to believe that the 0.3 μm particle is the one we should use for testing and we were told that the results we get with 0.3 μm particles are significant. What is the significance of 0.07 or 0.2 particles? Have you gone into that yet?

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SALTZMAN: If it is true that the size of maximum penetration is less than $0.3\ \mu\text{m}$, you are still doing adequate filter testing, even though you thought the aerosol was $0.3\ \mu\text{m}$. It seems to me that the instrumentation that was developed in the past has been adequate. The OWL was developed in the early forties. At some point, it is time to look at new instrumentation to give you a size distribution. The OWL does not do that. As you pointed out at a number of previous air cleaning conferences, the OWL is giving incorrect results if you need some way to measure size distribution. It makes sense to use a polydisperse aerosol to compute filter efficiencies as a function of size. If you have that information, that is all the information you are going to be able to get from a filter. I see no reason to continue using monodisperse aerosols. I think a polydisperse aerosol is more appropriate and if you obtain the size distribution upstream and downstream in some adequate fashion, that is the best information you are going to be able to get.

ANDERSON, W.L.: The Q107 is known to give a very high standard deviation, but it is also known that it can be controlled. Did you make any attempt to fine-tune the instrument in order to get that kind of data?

SALTZMAN: I am certainly no expert in operating the Q107. The test conditions were exactly those established in the daily operation of the filter test facilities by each of the operators at Oak Ridge, Hanford, and Rocky Flats. They ran the system and we sampled the aerosol. The aerosols were generated under what we took to be normal operating conditions, adjusted to $0.3\ \mu\text{m}$ with the owl.

FIRST: I would like to point out that a couple of conferences ago the Harvard Air Cleaning Laboratory published Q127 results made after the instruments had been fine-tuned. I think they were rather sharper and more on the beam than what you have observed in the filter test stations. I don't want to make any judgments as to the ability of the people who do this work but we did import an expert to get our machine working well.

ANDERSON: The Q127 can be fine-tuned. We have fine-tuned one to a standard deviation of 1.1. We still see a differential between sizes.

SALTZMAN: How did you determine the standard deviation?

ANDERSON: The standard deviation was determined by a whole series of different things: cascade impaction, mobility, and other types of measurements.

SALTZMAN: Have you published these data?

ANDERSON: We discussed the data with some of your people and I have it with me. I will be able to show it to you. The point I am trying to make is that the LAS-X is an excellent instrument for measuring aerosol particles of much greater size. It is only recently that it has been extended down into the lower size ranges. I am interested in knowing whether you are conducting any experiments to verify the claims of the manufacturer. For instance, coincidence

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counting. You indicated that there is no effect; yet, we see as much as 8% difference. We also are concerned about the fact that the LAS-X only counts about one particle out of 10. Is there any influence of particle size or particle concentration on the number of particles that the LAS-X displays?

SALTZMAN: If you are running it at an appropriate count rate, it counts every particle. One would want to run it at some reasonable count rate. They quote a maximum count at any given size range across either 15 or 16 channels, including the overflow channel, to be 25,000 counts per second. So, we don't have a problem of particles piling up with two particles in the sampling volume. You would have to run it at a very, very high concentration to get that. If you ran 10,000 particles a second over the whole range, I believe you could count all the particles. You might count only one in ten if you plugged the instrument directly into the output of the Q107, but, as I pointed out, we used the diluter.

ANDERSON: I use a different value, 5,000 particles per cc, whereas you reference it according to time. We are saying that you need a diluter to give 1,000 to 1 dilution rather than 500 to 1 as you have indicated.

SALTZMAN: I just picked 500 to 1. Our diluter operates from 200 to 1,100 to 1.

ANDERSON: I encourage you to obtain the rest of my data and examine it. Perhaps you will wish to modify some of the numbers you are using. I think the whole industry needs to have some kind of upper limitation in particle concentration, in time, and in volume flow through the system. I think what we are looking for from your organization is to straighten this whole picture out.

SALTZMAN: I appreciate that. I would, indeed, like to incorporate your data. As I mentioned, we are planning a collaborative study with an independent laboratory in Albuquerque to do comparisons using different types of instruments, in addition to the LAS-X, at the same site and at the same time to check out the instruments.

ANDERSON: What type of experiment were you anticipating that will verify the claims that have been advanced by the manufacturer? Have you used the LAS-X on other instruments that generate a more homogeneous aerosol?

SALTZMAN: Our present data confirm the manufacturer's claim of small dependence on refractive index. We have checked the LAS-X calibration with polystyrene latex aerosols that are quite monodisperse. We have obtained results for a monodisperse aerosol. We are planning a collaborative study with an independent group to compare the LAS-X with instruments such as the mobility analyzer.

MURROW: Have you tried the LAS-X on NaCl aerosol or has one been sent to England?

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SALTZMAN: We have not used the LAS-X at LANL with NaCl. However Pinnick and Ouvermann (J. Aerosol Science, Vol. 10, p 55-74 (1978)) have tested a related instrument with NaCl.

FIRST: Again, referring to the work that we did at HACL, we readjusted the Q127 until we got the aerosol to correspond to 0.03 μm diameter by the intercavity laser unit. Did you do any of these kinds of experiments to see if you could find a correspondence between the two instruments that would give you a constant ratio?

SALTZMAN: We did not. The objective was to look at the aerosols, as used, at the test stations rather than trying to force the OWL to agree with the LAS-X.

FIRST: The reason I bring it up is, if we are concerned that a particular size is the one we should be testing filters with because it is the minimum filterable size, then it would seem to me that one could calibrate the OWL by the laser spectrometer and continued to use the much simpler and cheaper instrument for routine testing. Has that been part of your considerations?

SALTZMAN: Your question suggests that the better approach might be to test with a polydisperse aerosol and use existing instrumentation to determine concentration and filter efficiency as a function of size (assuming the problem of dilution losses as a function of particle size can be eliminated or quantified). This would permit use of a simpler aerosol generation system and provide information regarding filter efficiency for the 0.3 μm aerosol assumed to be in use; or for an under 0.2 μm aerosol which apparently is actually used; or for any size or sizes of interest. This might provide both a simpler and also a more meaningful test. I agree that we should test with a polydisperse aerosol so that we can obtain filter efficiency as a function of particle size. With the instrumentation now in use at the filter test facilities (OWL and penetrometer) efficiency as a function of size cannot be obtained. A single particle spectrometer like the LAS-X will be needed to measure size distributions.

ETTINGER: The suggestion seems to be that possibly we should switch to a polydisperse aerosol. We don't know what is the size of maximum penetration since it varies with filter velocity and filter characteristics. If we pick any one size, assuming that we can generate a perfect monodisperse aerosol, it is a shifting target. By going to a polydisperse aerosol, which is generally easier to generate, and by going to new instrumentation which lets you calculate concentration as a function of size, assuming you solve the problem of dilution which Dr. Bergman mentioned, you can get all of the information in terms of what is the efficiency for every single size interval. Then you can say, "I am only interested in 0.3 μm to keep tradition with the past," or "I am only interested in 0.2 μm because that is what we really did in the past," or you can get any size-efficiency information you want. If I understand your comment, that might be a very logical step. I realized this is a big change and we don't make it rapidly, but it would be a logical next step to see whether we can give more meaningful information to the user, to the designer, to the environmentalists, and to the regulatory

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people by going to a simpler aerosol generation system using the LAS-X if it survives all the trials and tribulations and the multiple checks that we are trying to put it through. Thus, we will really know what the efficiency of the filter will be under any situations in the field.

FIRST: I agree that the comprehensive information of particle size-efficiency is very well worth knowing, but the question really comes down to what is being done at the filter testing stations, or at the manufacturers plant, with regard to the test? No matter what test we run, we don't have precise information on what the aerosol may be that the filter is going to encounter. So, having a particle-size efficiency spectrum may give more information than we really need or can use other than from a research standpoint. I am looking at the problem from the quality control standpoint. That is what the test was intended for. For a quality control test, one doesn't need to have large amounts of comprehensive information. I think there probably is a need for both kinds of measurements. In other words, an index of quality for routine testing and a much more detailed kind of test procedure for basic information about filters. I merely present this as a thought, not as a conviction of how we should go, because I think you will recall that I have advocated in the past (and I want to make sure that I am not hiding behind the wrong words here) eliminating the hot DOP test altogether for all except the paper and then simply using in-place testing for checking installation integrity, or assembly integrity. There are many ideas, and I think it is well that we consider them all to come up with what may be the best possible answer for the future.

GERBER: This is a question for Dr. First. Two conferences ago, in your paper on the particle size distribution from the Q107, you pointed out that the OWL was really showing a weighting to the seventh power. In light of that, how do you suppose that one could calibrate the instrument to eliminate that factor? Wouldn't it always show that weighted average and actually give you no information as to the amount of dispersity?

FIRST: I think your question gets at the root of much of what we are discussing in that, as soon as we change our particle sizing instrument to one which observes a different characteristic of the particles, we inevitably come up with a different number. We have a number of correlations which have been made in the past between salt aerosol and DOP, for example. It is not surprising, in my viewpoint at least, that if you use an intercavity laser you are going to get a different number than if you use an OWL or an electron microscope, or whatever. You are measuring a different characteristic of the particle and inevitably arrive at a different number. I don't know exactly how to reconcile this in terms of selecting the best possible instrument and the best possible test. It is sort of like going into the cafeteria and deciding which of the two dishes you would like for lunch.

SALTZMAN: In other words, what I am really saying is that the OWL, apparently, by your own words, is not unique. It gives an infinite number of answers all meaning 29 or 30 degrees on the meter. Twenty-nine or 30 degrees on the meter can be measured for an infinite

number of distributions.

FIRST: Assuming that the aerosol is not monodisperse, you are saying?

SALTZMAN: Yes. I think it is pretty clear that even a geometric standard deviation 1.1 is not a monodisperse aerosol.

FIRST: It depends on your definition of monodisperse. Our speaker quoted Fuchs. Now we have a new definition from you which says even 1.1 is not monodisperse. Where do you draw the line?

GERBER: Commenting on Dr. First's suggestion that the OWL could be calibrated with LAS-X and then used for quality control: How is this possible, given the fact that the OWL measures a weighted average (d^7 weighting) as published by Hines and First in the 1978 Nuclear Air Cleaning Conference? Due to non-uniqueness, one could never be confident of the circumstances being measured.

SALTZMAN: I believe the correct power was $d^{8.1}$. However, I do agree that a very large number of different distributions will give a mechanical analyzer reading of 29° , so it cannot be used to control the particle size.

Appendix E

OPERATIONAL EXPERIENCE USING DIETHYLHEXYLSEBACATE (DEHS)
AS A CHALLENGE TEST AEROSOL IN FILTER TESTING

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Abstract

Di(2-ethylhexyl)phthalate (DEHP) is the agent normally utilized to generate the challenge test aerosol for penetration testing of HEPA (High Efficiency Particulate) filters. At the Hanford Filter Test Facility (HFTF), di(2-ethylhexyl)sebacate (DEHS) has been used as a substitute for DEHP for over one year. The HFTF has conducted multiple studies to evaluate the performance of DEHS, as compared to DEHP, as a challenge test aerosol. Descriptive statistics are utilized to report the results of filters tested with:

- DEHP at HFTF and DEHS at HFTF
- DEHP at the Rocky Flats Filter Test Facility (RFFTF) and DEHS at HFTF
- DEHP by the manufacturer and DEHS at HFTF

Characterization of the DEHS aerosol, including particle size distribution and reproducibility, are described. In addition, operating temperatures, flow rates, concentrations, and preventive maintenance requirements associated with the use of DEHS at the HFTF are briefly presented.

Performance evaluations made by HFTF confirm that DEHS provides penetration test results equivalent to that obtained with DEHP. The experience obtained in the use of DEHS, at HFTF, supports its use as an acceptable substitute for DEHP in penetration testing of HEPA filters.

Introduction

During October, 1980, the National Toxicology Program released a draft carcinogenesis bioassay report on DEHP, indicating the substance was carcinogenic in two species of experimental animals.¹ Consistent with corporate and U.S. Department of Energy policies, the HFTF, operated by the Hanford Environmental Health Foundation (HEHF), implemented a program to reduce potential occupational exposure to DEHP. This included efforts to identify an alternative test aerosol agent equivalent in performance to DEHP.

Screening criteria were established by the HFTF for candidate materials for substitution to assure that aerosol agents selected for further evaluation could logically produce test results equivalent to that obtained by the commonly utilized test methods incorporating DEHP. The criteria included the following:

- Compatibility with existing facilities and equipment
- Exhibit behavior similar to DEHP using accepted test methods
- Effectively discriminate between penetration values of less than 0.03%
- Accurately replicate test results obtained with DEHP
- Able to reproduce test results on multiple testing

A review of the literature²⁻⁶ and consultation with aerosol physicists resulted in the selection of corn oil, polyethylene glycol (PEG 400) and DEHS as primary candidates for consideration. The criteria were best met by DEHS. Following evaluation and testing, the HFTF began using DEHS as a substitute test aerosol during January, 1981. Since that date, extensive efforts have been made by HFTF personnel in the evaluation of DEHS as an equivalent substitute for DEHP in quality assurance filter testing. Those studies of particular interest to organizations other than our own are presented.

Materials and Methods

The testing of filters incorporates many more variables than the testing of filter media only. This is attributed to the many components of filter construction including frame, gasket, filter pack, separators, and even the penetrometer systems with which the filters are tested. To minimize variables, filters for analysis were limited to 24 X 24 X 11 1/2 inch, 1000 cfm, standard design filters. For all tests, filters were inspected, tested, labeled, and data recorded in the usual manner. Immediately prior to repacking the filters, the manufacturer's or other appropriate test data were also recorded.

The filters used for data collection in this study were not selected using statistical sampling methods. In every case, the filters represent 'convenience' samples from the U.S. Department of Energy's Hanford facility stock piles. Available inventory was the usual determinant of sample size.

Physical Properties of DEHS

Table 1 summarizes the physical properties of DEHS as compared to DEHP. Chemically, DEHS and DEHP are very similar; however, DEHP contains an aromatic ring and in DEHS the corresponding structure is a saturated aliphatic. Some reviewers have suggested this may account for differing biological response.⁷ The similar physical properties suggested comparable performance in penetrometer applications.

Table I. Physical properties of DEHP and DEHS

	<u>DEHP</u>	<u>DEHS</u>
Chemical Formula	C ₂₄ H ₃₈ O ₄	C ₂₆ H ₅₀ O ₄
Elemental Composition (percent)		
Carbon	73.80	73.19
Hydrogen	9.81	11.81
Oxygen	16.39	15.00
Molecular Weight	390.54	426.66
Specific Gravity (@ 25°C)	0.982	0.913
Index of Refraction (@ 25°C)	1.485	1.449
Vapor Pressure (mm Hg @ 25°C)	<1	<1
Boiling Point (°C @ 4 mm Hg)	222-230	235-248
Flash Point (°F/COC)	425	430
Viscosity (@ 25°C cps)	57	25
Cost (\$ per lb)	0.47	1.98

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Table II characterizes typical particle size distributions for test aerosols at the U.S. Department of Energy Filter Test Facilities (FTF). The size distributions were all recorded utilizing an LAS-X spectrophotometer⁸ and corrected for differing refractive indices of the aerosol agents. The particle distributions were discussed in detail in a paper presented earlier at this conference.⁹

Table II. Particle size characterization of challenge test aerosols used at the FTF's*

	Geometric Mean (\bar{X}_g) in μm	Geometric Standard Deviation (S.D. _g) in μm
Rocky Flats (DEHP)	0.192	1.451
Oak Ridge (DEHP)	0.180	1.380
Hanford (DEHS)	0.173	1.345

* Data expressed as count mean diameter (cmd)

Operating Conditions

Normal HFTF operating conditions using DEHS as a challenge test aerosol in the Q107 penetrometer¹⁰ are provided in Table III. Similar data for DEHP is provided for comparison purposes.

Table III. Normal operating conditions using DEHS vs DEHP at HFTF (Q107 Penetrometer)

<u>Parameter</u>	<u>DEHS</u>	<u>DEHP</u>
Quench temperature (°F)	114-120	92-110
Liquid temperature in reservoir (°F)	364-400	389-400
Vapor temperature above reservoir (°F)	292-327	274-290
Vapor flow (cfm)	45-60	50-65
Quench flow (cfm)	110-205	145-170
Main flow (cfm)	980-1360	980-1140
Aerosol concentration ($\mu\text{g/L}$)	45-64	46-77

Preventive Maintenance

In conjunction with the initial operational testing of DEHS as a candidate aerosol agent, a tar-like residual was observed on the liquid reservoir immersion heaters of the HFTF Q76 Penetrometer. The routine preventive maintenance program was modified to include draining 'used' DEHS after five days of equipment operation. The residual has not since been observed. Differential thermal analysis of samples of DEHP and DEHS did not indicate significant differences in thermal degradation patterns. The initial residues may have been due, in part, to residuals or complexes of other compounds tested including corn oil and PEG 400.

An in-house material certification test has also been initiated. Samples are collected from each lot (drum) of DEHS received and the index of refraction and specific gravity are measured. In addition, an infrared scan is made to determine if gross impurities are present.

DEHS vs DEHPComparability of Individual Measurements

Fifty filters were located that had been previously tested (August, 1980) by the HFTF with DEHP and were retested at the HFTF (February, 1981) with DEHS. The results of this retest are depicted in Figure 1. The initial DEHP values are indicated by circles and subsequent DEHS test results are indicated by squares. As summarized in Table IV, utilizing standard tests at 100% and at 20% of the manufacturer's rated airflow, seven of the 50 filters retested were observed to differ from the initial test values by more than 10 percent of the reference value (0.03% penetration).

These differences were noted in all seven filters at the 20% flow rate and only five of the seven at the 100% flow rate. At 20% of the rated flow, four of the seven had differences greater than 25% of the reference value. Only two of these four had differences greater than 33% of the reference value. We chose to examine more closely those filters which retested with differences greater than 33% of the reference value to see if we could identify the reasons for the differences.

Subsequent visual examination of both filters revealed pinhole leaks in the media or gasket voids attributable to local handling damage. For purposes of this study only, repairs were attempted to determine the sources of the differences. Repair of the obviously damaged filters resulted in values agreeing within 10% ($\pm 0.003\%$ penetration) of the original data obtained with DEHP.

The most important factor to be considered when testing with a substitute aerosol is whether use of the substitute would lead to the acceptance of filters that would otherwise be rejected. The data presented in Table IV does not support such a conclusion. Similarly, the DEHS test method does not result in rejecting filters that would otherwise be accepted with DEHP. The two filters rejected with repeated DEHS testing were found to be damaged.

Table V summarizes the distribution of test data obtained for filters tested with DEHP and DEHS at the HFTF.

Table V. Distribution of test data for filters tested with DEHP and DEHS at HFTF

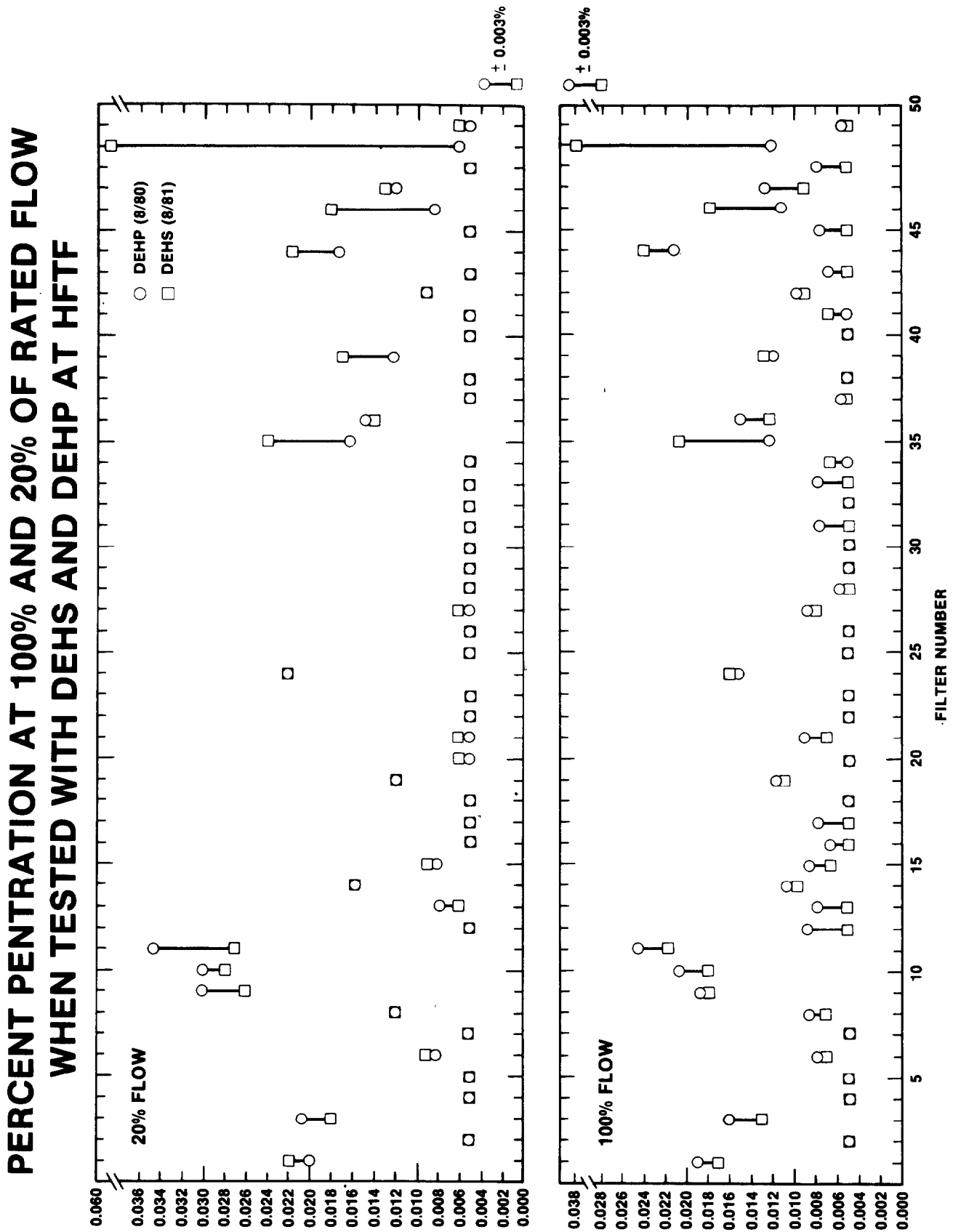
Test Condition	% Penetration Including Damaged Filters (N = 50)		% Penetration Excluding Damaged Filters (N = 48)	
	\bar{x}	S.D.	\bar{x}	S.D.
DEHP/20% Flow	0.009	0.007	0.009	0.007
DEHS/20% Flow	0.011	0.010	0.009	0.007
DEHP/100% Flow	0.009	0.005	0.009	0.005
DEHS/100% Flow	0.009	0.007	0.008	0.005

Table IV. Observed differences in detected penetration values for DEHP and DEHS at HFTF.

Test Condition (Reference 1000 cfm)	Absolute Difference*			Rejected, Previously Accepted
	>0.003 (10%)	>0.007 (25%)	>0.010 (33%)	
20% Flow	7	4	2	2
100% Flow	5	2	1	1
Total	7/50	4/50	2/50	2/50

* Approximately 10%, 25%, and 33% of the test criteria for rejection.

FIGURE 1



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The observed mean value and standard deviation clearly indicate the homogeneity of the test data obtained with either aerosol agent. Assuming DEHP measures the true penetration value, DEHS is equivalent to DEHP in discriminating at less than 0.03% (the accepted criteria for rejection of HEPA filters). This reflects the ability of DEHS to achieve equivalent decisions, regarding acceptance or rejection, as DEHP.

Comparability Between FTF's

Figure 2 illustrates HFTF test data as compared to that obtained from RFFTF. The HFTF does report slightly higher penetration values than those obtained at the RFFTF with about the same relative variability. The difference in the means may be due in part to a slightly smaller particle size obtained with DEHS (note Table II). However, Figure 3 illustrates the comparison of the HFTF test results obtained with DEHS as compared with the manufacturer's test data obtained with DEHP. The means of the test data obtained by the HFTF and manufacturer are in very close agreement. Table VI is a summary of the variability of test measurements made on identical filters by RFFTF and the manufacturer using DEHP, and HFTF using DEHS. From the coefficients of variation, it appears that the total variability of the test method, including aerosol agents, equipment, and personnel, were essentially constant. The test facility, including aerosol agents, may be chosen without concern.

Table VI. Means, standard deviations and coefficient of variation for filter test data on identical filters.

	\bar{X}	S.D.	$CV = \frac{S.D.}{\bar{X}}$
RFFTF (DEHP)	0.0085	0.0023	27%
MFG (DEHP)	0.0142	0.0044	31%
HFTF (DEHS)	0.0113	0.0038	34%

Figure 4 illustrates the distribution of test results obtained on over 100 filters tested by HFTF in August 1981, February 1982, and April 1982. The number of filters vary over time due to shipment damage or unavailability of test results. These data clearly indicate the reliability of DEHS for obtaining reproducible results over time.

Discussion

As previously stated, the testing of filters incorporates many more variables than the testing of filter media only. These variables represent the many components of filter construction including frame, gasket, and even the penetrometer systems with which the filters are tested. For example, at a time when DEHP was used for filter testing by HFTF and similar standard filters tested, variability in test data existed (Figure 5). This observation is independent of the choice of aerosol agent. This is attributed to other variables in testing such as equipment, personnel, and laboratory environments.

Figures 3 and 5 also illustrate a noteworthy observation frequently made with filter test data. An obvious digit preference is indicated as odd digits were rarely recorded by the manufacturer. This may well have resulted in a systematic bias of all penetration values.

FIGURE 2

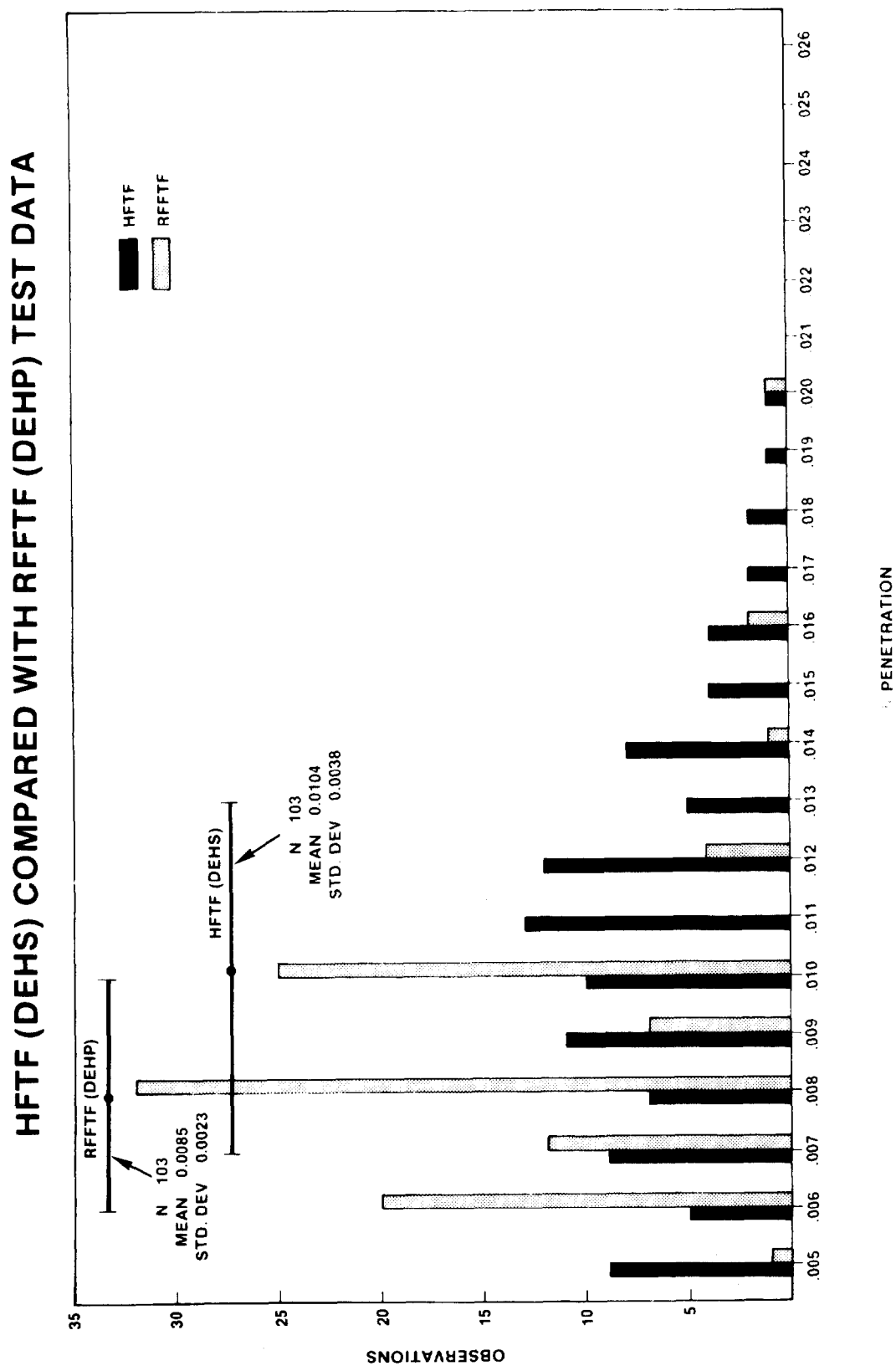


FIGURE 3

HFTF (DEHS) COMPARED WITH MANUFACTURER'S (DEHP) TEST DATA

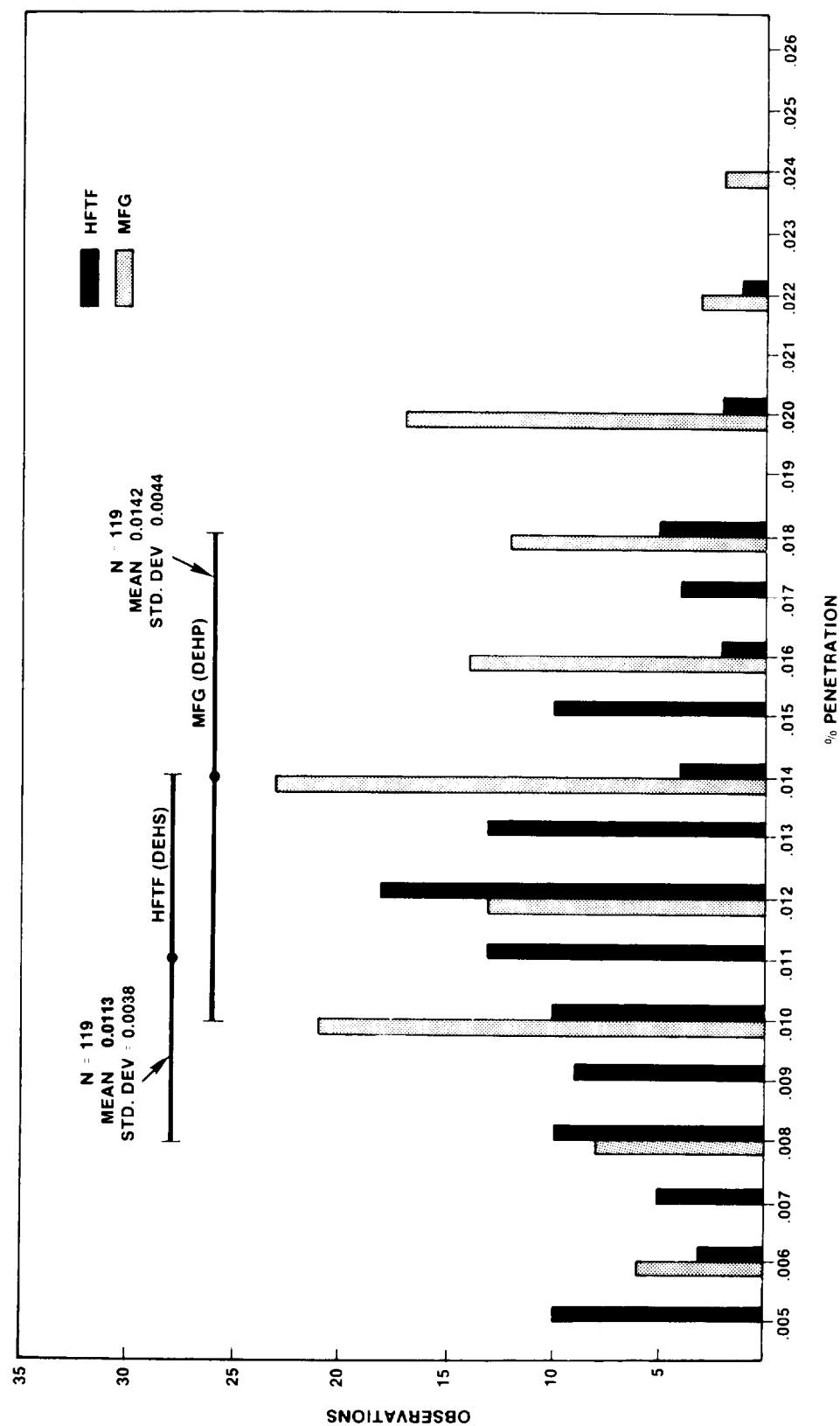


FIGURE 4

PRECISION OF DEHS TEST DATA ILLUSTRATED BY FILTER TESTS ON THREE SEPARATE OCCASIONS

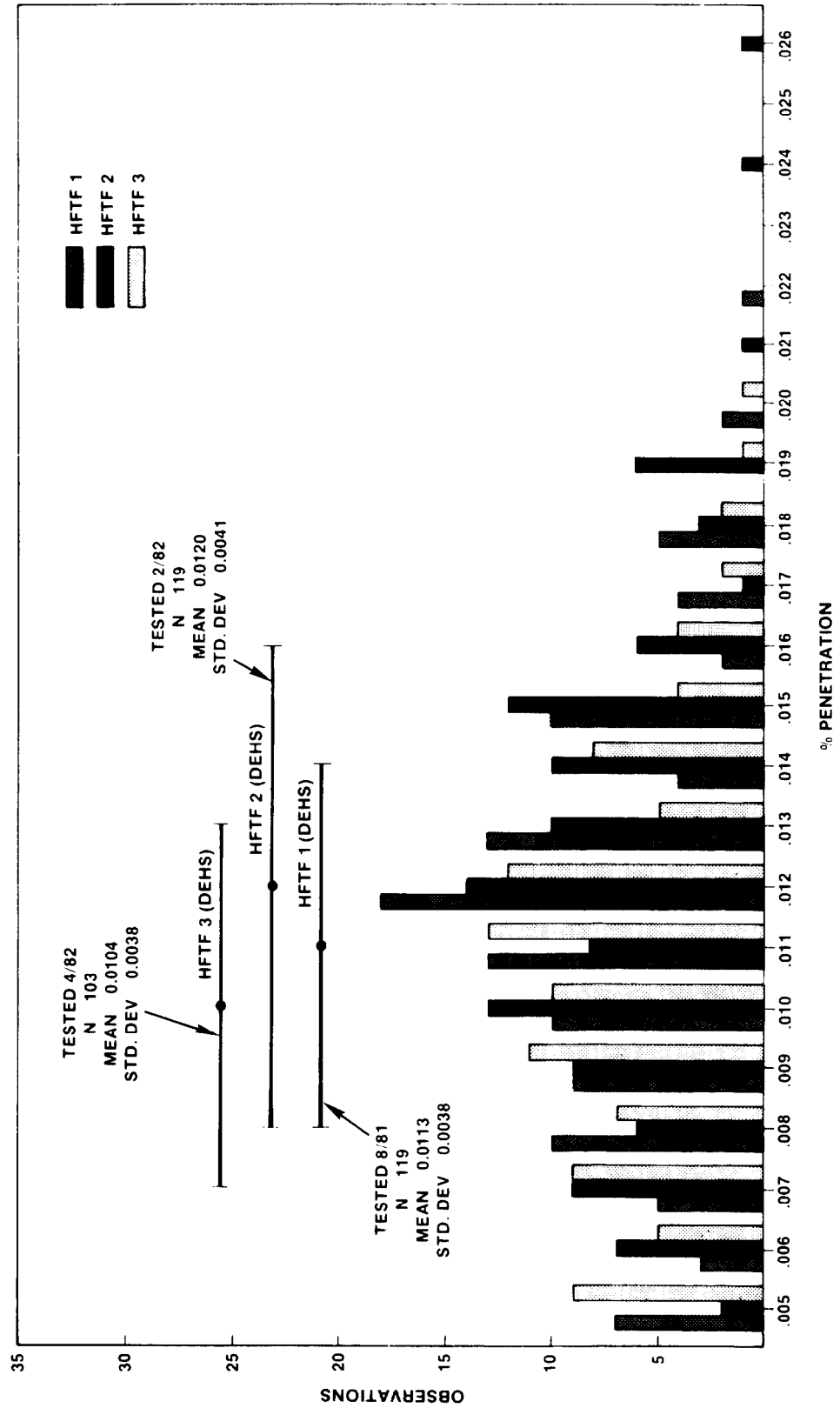
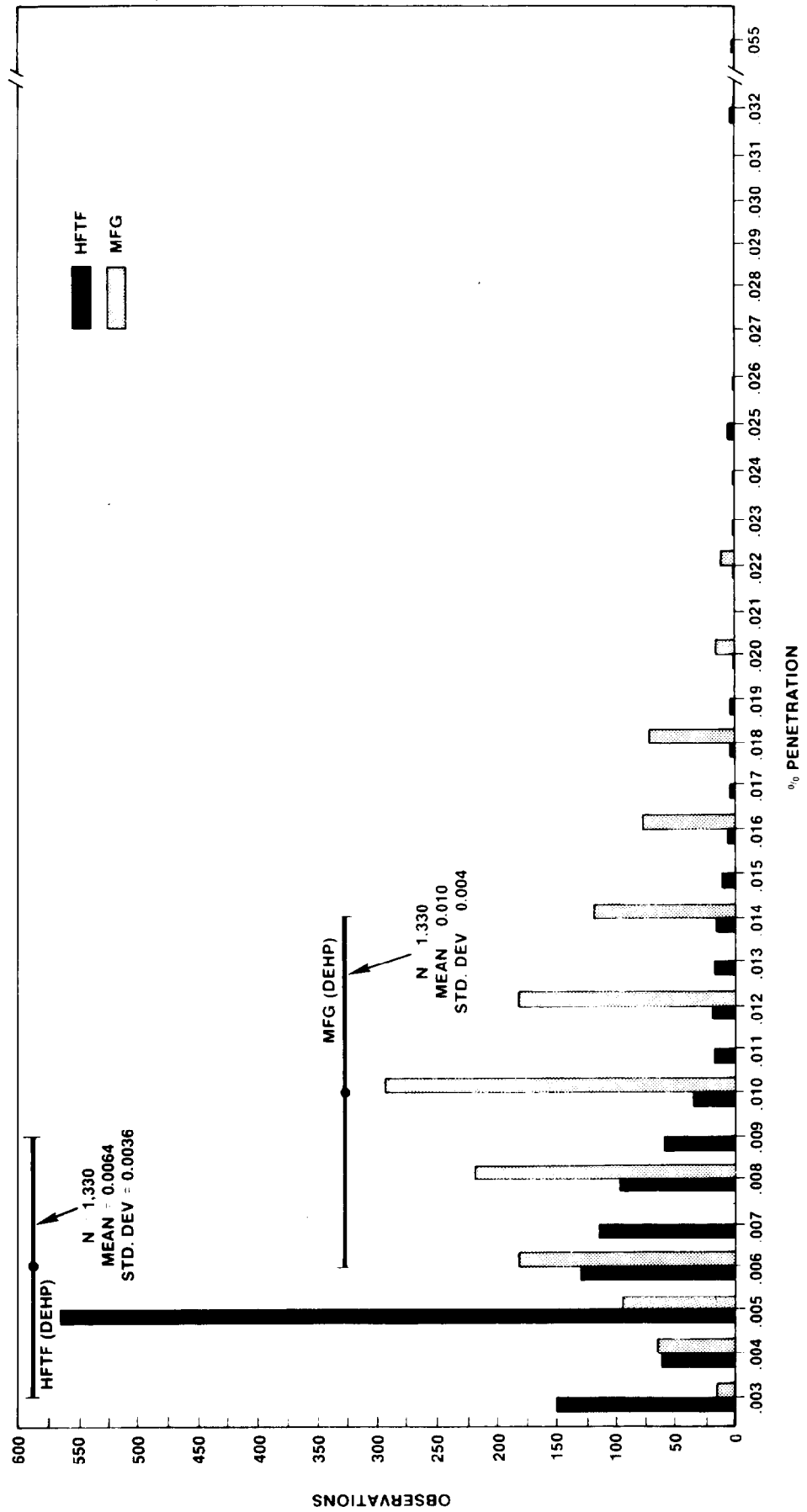


FIGURE 5

HFTF (DEHP) COMPARED WITH MANUFACTURER'S (DEHP) TEST DATA



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Variation is inherent in all operational data. This variability is seen in all filter testing data, including the data reported in this study. We suggest that using mean values (point estimates) without including a measure of inherent variability (such as the standard deviation) is inappropriate. For purposes of our review of the data, we simply chose to look at the mean plus or minus one standard deviation. This is not a statistical test. However, it has shown descriptively that the mean values are very likely to be the same when this measure of variability is included. This is analogous to conducting more formal testing using confidence intervals.

Conclusions

The following conclusions are reached from the data presented:

- DEHS is compatible with the commonly used filter testing facilities and equipment
- DEHS exhibits similar operational behavior to DEHP using accepted test methods
- DEHS effectively discriminates between penetration values of less than 0.03%
- DEHS accurately replicates test results obtained with DEHP
- DEHS exhibits relative precision as indicated by similar coefficients of variation

Therefore, DEHS is an acceptable substitute challenge aerosol in HEPA filter testing.

Acknowledgements

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DISCUSSION

THOMAS, T.R.: Your standards committee is writing standards to make testing procedures at the DOE Filter Test Stations more uniform. Do you see any problem with HEAF using DOS, and the other two stations using DOP?

BURCHSTED: It is premature to make a response to this point until it is discussed in committee. However, it does appear to be inconsistent for the QA Filter Test Facilities to be using two aerosols

FIRST: I believe the sense of the paper we just heard was that it makes no difference whether you use one or the other, in terms of filter testing, although, I think it is obvious that before a standard is promulgated, it would be nice to have at least one other laboratory confirm the results.

GILMORE: I would like to point out that I have data on almost 5,000 filters. There is no greater variability using DEHS than there is using any other filter test method. The basis of our presentation today is that the aerosol agents are, indeed, equivalent, and the variability in using one is no greater than with any other. I think that variability is an issue that needs to be addressed and should be the subject for further research by all the filter test facilities. That is the only issue that needs to be addressed.

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THOMAS, T.R.: EPA has decided not to ban industrial and commercial uses of DOP. In light of this, will you consider lifting your ban.

GILMORE: The EPA, or the National Institutes of Health, have not reversed their opinions that DEHP has been shown to be carcinogenic in two species of animals. The EPA has only chosen not to regulate DEHP as a priority chemical. The decision to seek a substitute for DEHP was based on its potential to be carcinogenic. Until evidence to the contrary is firmly established, there is no basis for reversing our position.

STEINBERG: It is true that there was an article in the San Francisco Chronicle on January 12 that said DOP is no longer considered a carcinogen.

ETTINGER: Replying to Mr. Steinberg, EPA did not claim that DEHP was not a carcinogen. They said that there was insufficient evidence to regulate DEHP as a priority chemical. This is the same stand that they took relative to formaldehyde. This is not the same as saying DEHP is not a carcinogen.

ANDERSON, W.L.: Since the ethyl-hexyl radical is the potential carcinogen and this is common to both DOS and DOP, the selection should be based upon other physical characteristics of the test aerosol.

GILMORE: We are aware that some individuals have suggested 2-ethyl hexane may be a biologically active compound. However, it has not been confirmed as the active compound in DEHP carcinogenesis. Our review of the literature, and consultation with toxicologists, confirms that DEHS is a reasonable compound to consider for substitutes for DEHP. DEHS is certainly the compound that is most similar in terms of other physical properties.

KOVACH, J.L.: The only common property demonstrated is that neither DOP nor DOS generates 0.3 μm aerosols as used at the FTFs. Also the use of a laser instrument will not correct the polydispersity of the aerosol used.

GILMORE: DOP and DOS are currently producing the same aerosol for filter testing that they always have. We may have erroneously assumed it to be a 0.3 μm aerosol, but it remains the common, reproducible quality assurance test it always has been. The current issue should be what criteria do we really want for aerosol testing, i.e., particle size, size distribution, etc. Is the current DOP method really the best or is it even adequate? The operational experience would suggest it is certainly adequate. However, the laser offers the potential of providing additional information which may be very useful.

FIRST: I believe Dr. Saltzman said the filter test stations are not able to maintain a consistent aerosol from time to time. If I am quoting him correctly, I think that would indicate that your statement that the aerosol is the same today as it was in 1941 doesn't necessarily hold true. It may be quite variable if the test stations do not generate exactly the same aerosol from one month, or one year,

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to the next.

GILMORE: I concur with you. The difference is that the filter test stations are operated twelve months out of the year and not for a specific period of time or for a specific research project. There is variability of laboratory conditions and environmental conditions and personnel change frequently. Those are all true variables in the generation of data. Indeed, particle size will vary as a function of all of the variables. These are the same types of variables (and manufacturers concur) that we have been testing with since 1941. If you accept the inherent variables which we illustrated in our paper, the aerosols are the same now as they were in 1941.

SALTZMAN: In response to Dr. First's comment, I don't believe that I said the aerosol size distributions at the FTFs varied from month to month. In fact, they consistently generated an aerosol with the same broad polydisperse distribution.

Appendix F

THE NEED FOR REVISING
U.S. NUCLEAR REGULATORY COMMISSION REGULATORY GUIDE 1.52
IN LIGHT OF NRC-SPONSORED RESEARCH PROGRAM RESULTS
AND OTHER DEVELOPMENTS

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ABSTRACT

Since the previous revision of Regulatory Guide 1.52, Design, Testing and Maintenance Criteria for Post Accident Engineered-Safety-Feature Atmosphere Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants, made in 1978, a number of events have occurred or are underway which will result in a schedule being established for the next revision of this Regulatory Guide. The results of U.S. Nuclear Regulatory Commission (NRC) sponsored research at the U.S. Naval Research Laboratory on the Effects of Weathering on Impregnated Charcoal Performance and Charcoal Performance Under Accident Conditions are important factors which contribute to the need for revision. This research has shown that the type of impregnant greatly influences the weathering characteristics of the charcoal and that at least some weathered charcoals show improved performance after radiation exposures in the range of 10^7 to 10^9 rads.

Several other factors are contributing to the need for revision and will influence the subsequent timetable for revision. Among these factors are: a) NRC endorsement, in whole or in part, of ANSI/ASME N509-1980 and ANSI/ASME N510-1980 needs to be accomplished; b) questions to the NRC have been raised addressing the appropriateness of its present position of not requiring HEPA filter testing at Department of Energy Quality Assurance Testing Stations; and c) post TMI-2 accident-related concerns.

The paper discusses the need for initiating another revision to Regulatory Guide 1.52 and identifies preliminary positions on certain issues (endorsement of N509/N510, e.g.) that are appropriate for consideration in the draft of a proposed Revision 3 to Regulatory Guide 1.52.

Introduction

General Design Criteria 19, 41, 42, 43 and 61 of Appendix A, "General Design Criteria for Nuclear Power Plants," to 10 CFR Part 50, "Licensing of Production and Utilization Facilities," require that adequate radiation protection be provided to permit access to and occupancy of the control room under accident conditions; that containment atmosphere cleanup systems be provided, as necessary, to reduce the amount of radioactive material released to the environment following a postulated design basis accident (DBA); that fuel storage and handling systems, radioactive waste systems, and other systems that may contain radioactivity be designed to ensure adequate safety under postulated accident conditions and that they be designed with appropriate containment, confinement, and filtering systems; and that engineered safety feature (ESF) filtration systems be designed to permit appropriate periodic inspection and testing to ensure their integrity, capability, and operability.

Regulatory Guide 1.52, Revision 2, March 1978,⁽¹⁾ specifies the methods that are presently acceptable to the U. S. Nuclear Regulatory Commission (NRC) staff for implementing the NRC's regulations in Appendix A to 10 CFR Part 50 with regard to design, testing, and maintenance criteria for air filtration and adsorption units of ESF atmosphere cleanup systems in light-water-cooled nuclear power plants. Since the publication of Rev. 2 to this Regulatory Guide, significant developments have occurred (e.g., ANSI/ASME N509 and N510 revision, TMI-2 accident) or are underway (NRC sponsored research on charcoal adsorber performance under simulated accident conditions and the chemical forms of radioiodine to be expected after an accident) which call for another updating in the not too distant future. The remainder of this paper will discuss these events and their potential impact on the next revision to Regulatory Guide 1.52.

Discussion

ANSI/ASME N509-1980⁽²⁾ and ANSI/ASME N510-1980⁽³⁾

At the time of the preparation of Revision 2 to Regulatory Guide 1.52 in 1978, both ANSI/ASME N509-1976⁽⁴⁾ and ANSI/ASME N510-1975⁽⁵⁾ were under revision. The decision was made at that time not to delay the publication of Revision 2 by waiting until N509-1976 and N510-1975 were revised. Rather, it was planned that a future Revision 3 to Regulatory Guide 1.52 would address the appropriateness of endorsing the updated N509 and N510 standards.

The publication of several revisions to Regulatory Guide 1.52 since the mid-1970s and the 1980 revisions to N509 and N510, however, have contributed to uncertainty in the U.S. regarding both the most appropriate and legally required codes/standards/procedures to be used in the qualification and testing of nuclear power plant filtration system components. For example, existing nuclear power plant licenses issued by the NRC either have unique surveillance requirements for ESF filtration system testing, a requirement for testing with reference to N510-1975, or a requirement for testing with reference to N510-1980. To promote consistency among the ESF filtration system surveillance requirements issued to different nuclear power plants and to ensure that filtration systems are likely to be as effective for removal of particulates and radioiodine as assumed in the NRC accident analyses, the NRC is completing a multi-plant activity that will

result in all operating nuclear power plants having technical specifications which require surveillance testing of ESF filtration systems consistent with at least the intent of Regulatory Guide 1.52.

Since the publication of N509-1980 and N510-1980, the NRC has received numerous inquiries questioning whether the NRC endorses the 1980 revisions and whether nuclear power plant applicants and licensees can use the 1980 versions even though their existing NRC license or commitments made to the NRC refer to N509-1976 and N510-1975. Although official endorsement of N509-1980 and N510-1980, in full or in part, will not occur until the next revision to Regulatory Guide 1.52 is completed, the NRC has generally held that use of the 1980 versions of the standards is acceptable when questioned by individual applicants, licensees, or filter vendors and testing contractors. The NRC has also generally held the position that once an applicant commits to a filtration system design in accordance with codes, guides and standards in existence at the time of the commitment, the applicant is not required to modify the design of the system as revisions to codes, guides and standards occur.

To a certain extent, periodic revisions to codes, guides and standards will necessarily create inconsistency and some confusion in regard to ESF filtration system design and surveillance testing requirements of the NRC. The next revision to Regulatory Guide 1.52, which will address N509-1980 and N510-1980, will clarify NRC's intent regarding the endorsement of those standards and promote understanding throughout the industry.

NRC-Sponsored Research on In-Service Charcoal Performance Under Simulated Accident Conditions

Following the accident at TMI-2, the NRC contracted with the U.S. Naval Research Laboratory (NRL) (Victor R. Deitz, investigator) to conduct a research program to study the combined effects of in-service weathering, exposure to atmospheric contaminants, and exposure to radiation on the retention of radioiodine by various nuclear-grade carbons. The first interim report⁽⁸⁾ on this research has been published and the results will be reported by the investigator at this conference. It is possible that when this research program is completed, some revision to the radiation level environmental design criteria for radioiodine buildup on charcoal adsorbers may be called for. The present position in Rev. 2 of Regulatory Guide 1.52 calls for qualification of the charcoal adsorbers in ESF atmosphere cleanup systems to design basis accident (DBA) conditions to a dose of 10^9 rads resulting from radio-iodine buildup on the adsorber. Based on the preliminary findings of this research program, absorbed dose to certain types of weathered charcoal after a DBA may be of less concern.

The NRL research program is exposing a number of service and weathered carbons (KI, TEDA, KI + TEDA impregnations) to gamma radiation from a Co-60 irradiation source and to the radiation from a linear accelerator (LINAC). Exposures of 10^6 , 10^7 , 10^8 and 10^9 rads are made and the methyl iodide-131 removal efficiency measured after each exposure.

In the future, this research program will continue to evaluate the effects of radiation exposure on methyl iodide penetration of various types of weathered charcoal samples. Irradiation of the charcoals will be conducted under no-flow

(static) conditions and in air flows containing different iodine concentrations. The iodine species desorbed from the charcoal adsorber will also be identified. The results from this research, as indicated previously, will be evaluated by the NRC and appropriate changes will be incorporated in future revisions to Regulatory Guide 1.52.

Impact of Nuclear Reactor Accident Source Term Assumptions

Analyses of the TMI-2 accident of March 1979 revealed that while between 3.5 and 13 million Curies of xenon escaped to the environment, it is estimated that only 13 to 18 Curies of radioiodine were released. This apparent disparity has rekindled concern about the appropriateness of current NRC assumptions regarding the amount and chemical form of fission product iodine available for release after an accident and the potential for releasing significant amounts of aerosols to the environment. This concern has been expressed in a variety of forms including: written communications⁽⁷⁾ to the Chairman of the U. S. Nuclear Regulatory Commission; presentations before the Commission on November 18, 1980; presentations⁽⁸⁾ to the Nuclear Safety Oversight Committee of the U. S. Congress; and papers^(9, 10) presented at professional society meetings.

The basis for the concern that was expressed to the NRC is the belief that, in most reactor accident sequences, iodine emerges from the fuel as cesium iodide and that the accident environment would sustain this chemical state. Further, it is believed that the accident environment would convert other iodine species be present to the less volatile iodide state. At about 400 C the cesium iodide would condense and plate-out on metal surfaces and eventually enter into solution as the iodide ion once water was encountered. This belief was the basis for the concern that iodine volatility assumed by existing NRC Regulatory Guides^(11, 12) was greatly overstated and that the radiological consequences of some nuclear accidents may be substantially less than those assumed at present.

Research is being sponsored by the NRC to investigate radioiodine volatility during and after an accident and the formation of aerosols. A number of specific regulatory requirements or guidance could be affected by substantive changes in the assumptions concerning accident source terms. As new information is obtained by the NRC that should be incorporated into its accident source term assumptions, requirements and guidance in the areas of emergency planning, siting, minimum engineered safety features, and degraded core may be revised. The requirements or guidance that may be affected by potential changes to accident source term assumptions include:

1. Regulations (10 CFR Parts 50 and 100).
2. Regulatory Guides 1.3, 1.4, 1.5, 1.7, 1.25, 1.52, 1.77, 1.96, 1.97, 1.145, and 4.7 (Refs. 11-15, 1, 16-20).
3. Emergency Preparedness Procedures.
4. Methods for Assessing Accident Environmental Impact.

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Regulatory Guide 1.52 (Rev. 2) specifies typical environmental accident conditions to be used for ESF primary and secondary atmosphere cleanup systems design. The specified values of radioiodine buildup on adsorbers and the airborne concentrations of elemental iodine, methyl iodide and particulate iodine are based on source terms specified in Regulatory Guides 1.3 or 1.4 (25% of core iodine inventory available for leakage from containment). In the event new accident source term assumptions are adopted, the ESF filtration system design capacities will need revision. One change to Regulatory Guide 1.52 would likely involve a new assumption about an increased percentage of aerosols in the containment atmosphere versus the present assumptions which dictate that filtration system design be based on the removal of volatile forms of iodine. Prefilter and HEPA filter design requirements will also need to be reexamined based on revised fission product source term and chemical form assumptions, especially the loading factor of aerosol postulated for selected accident sequences in the containment atmosphere.

Additional Candidate Topics or Positions for Revision in Regulatory Guide 1.52

Filtration experts have held informal discussions with the NRC expressing concern about certain regulatory positions contained in Rev. 2 to Regulatory Guide 1.52. The first concern relates to testing of HEPA filters at U. S. Department of Energy (DOE) Quality Assurance (QA) Testing Stations. In Revision 1 to Regulatory Guide 1.52, issued in July 1976, the NRC recommended that HEPA filters for use in ESF atmosphere cleanup systems be visually inspected and dioctyl phthalate (DOP) tested at DOE QA Filter Testing Stations prior to their installation and use in commercial nuclear power plants. This recommendation was predicated on the practice initiated by DOE and its predecessor agency in the early 1960's and remains in effect today for all HEPA filters designated for use in DOE facilities. In the preparation of Revision 2 to Regulatory Guide 1.52, an analysis was performed of the test results obtained by the DOE-QA Filter Testing Stations during the period 1971 to 1977. The results⁽²¹⁾ of this evaluation indicated that the filter rejection rate was low and the recommendation for HEPA testing at DOE QA Filter Testing Station was not included in Revision 2. As reported by Burchsted⁽²²⁾ at this conference, it appears, however, that while more recent data from the DOE QA Filter Testing Stations show no increase above prior years in HEPA filter rejection for reasons of excessive penetration, filter rejection for causes related to manufacturer quality control/quality assurance (pressure drop, loose gaskets, physical damage from handling) may have increased significantly. The next revision to Regulatory Guide 1.52 will reevaluate the present position that HEPA filter testing at DOE-QA Filter Testing Station is not required.

Questions have also been raised informally about the Regulatory Guide 1.52 positions which recommend that the volumetric air flow rate of a single cleanup train should be limited to 30,000 cfm and that laboratory testing of representative samples of in-service charcoal adsorbers be conducted after each 720 hours of system operation. Certain organizations which conduct in-place filter tests now claim that concern about the ability of equipment to generate sufficient amounts of test aerosol to leak test systems with flows significantly in excess of 30,000 cfm are unwarranted. Other industry representatives have informally voiced concern that the laboratory testing of charcoal adsorber samples after

each 720 hours of operation imposes too great a testing burden for plants which have ESF filtration systems that also operate frequently during normal operating conditions. Both of those regulatory positions will also be reevaluated in the next revision to Regulatory Guide 1.52.

One presently identified issue that will need evaluation for possible incorporation into the next revision of Regulatory Guide 1.52 remains. This issue was identified as a result of another research program sponsored by the NRC at the U. S. Naval Research Laboratory (Victor R. Deitz, principal investigator). This program investigated the effects of weathering on impregnated charcoal performance under conditions of normal operation. The results⁽²³⁾ of this research indicate that carbons with certain impregnation formulations (TEDA or TEDA + KI) exhibit less penetration of methyl iodide-131 when exposed to contaminants in outdoor air than do carbons impregnated with only KI and I. See Figure 1. Thus, in the next revision to Regulatory Guide 1.52 the NRC must decide whether it is appropriate to include guidance on acceptable charcoal weathering characteristics under certain assumed environmental operating conditions.

Summary and Conclusion

Regulatory Guide 1.52, "Design, Testing and Maintenance Criteria for Post-Accident Engineered-Safety-Feature Atmosphere Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants," was last updated as Revision 2 in March 1978. Since that time, additional research has been conducted that may potentially impact atmosphere cleanup system design guidance endorsed by the NRC. The impetus for much of this research was the accident at TMI-2 in 1979 and the subsequent reevaluation by the NRC of the assumptions used in accident source term calculations, especially as these assumptions relate to the relative abundance of iodine species available for release to the environment. Other developments such as publication of the 1980 revisions to ANSI/ASME N509 and N510, advances in technology, and additional experience obtained by the industry have also occurred since March 1978. The NRC presently anticipates that efforts to draft Revision 3 to Regulatory Guide 1.52 will commence in fiscal year 1983, beginning October 1, 1982. Members of industry are invited to identify candidate issues for incorporation into the revision and to respond when draft Revision 3 is published for comment. To the extent consistent with the safety responsibilities of the NRC, Revision 3 will continue to endorse consensus standards developed by air cleaning technical experts.

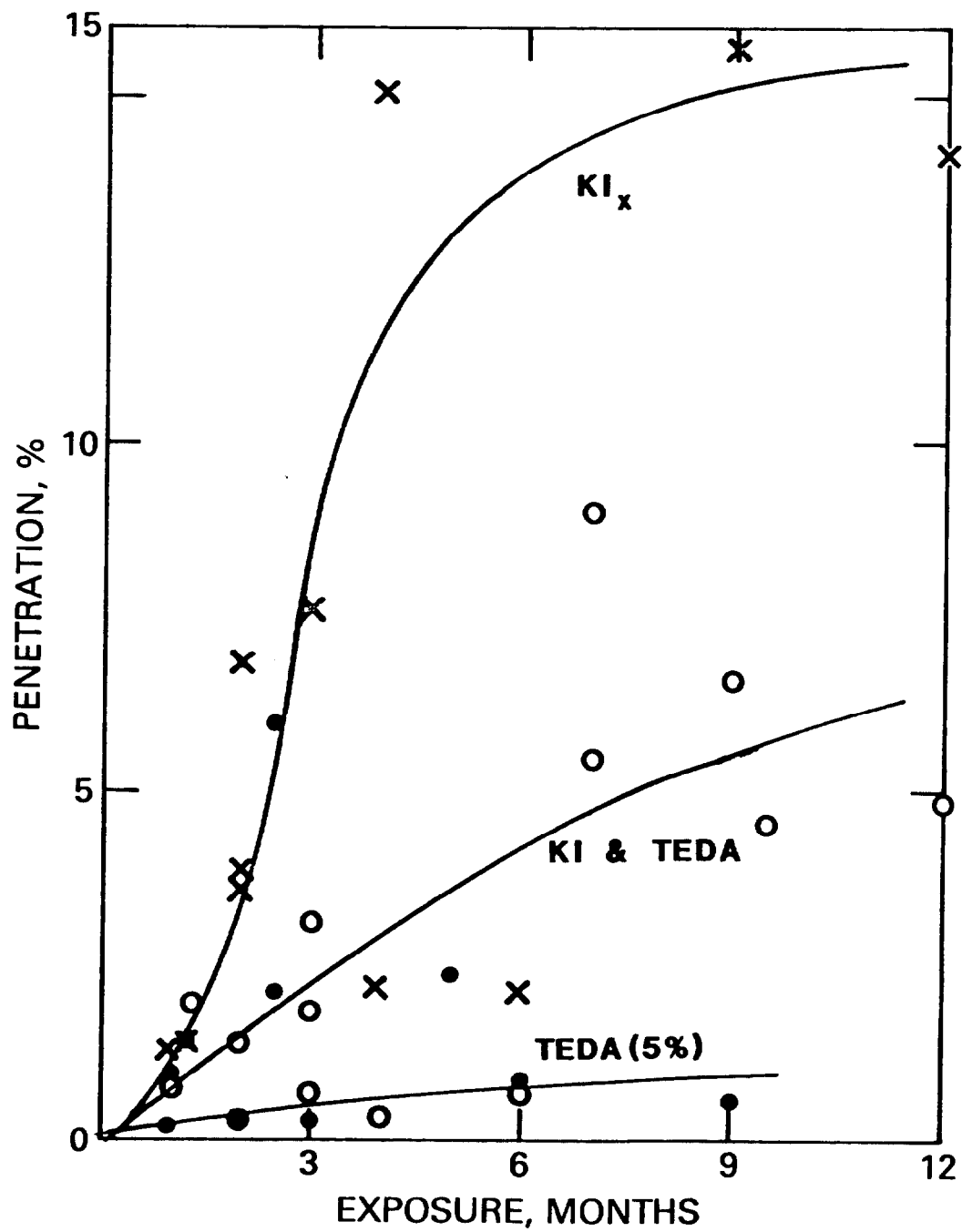


FIGURE 1
PENETRATIONS OF $^{131}\text{ICH}_3$ AFTER
EXPOSURE TO OUTDOOR AIR⁽²³⁾

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DISCUSSION

FIRST: You have projected the issuance of a revised RG 1.52 three years into the future. Does that seem like a reasonable estimate to you, Mr. Bangart?

BANGART: Hopefully not three years, but certainly, greater than one year. One and a half to two years is a good target for us to shoot for.

FIRST: Is that a usual time span? It seems a rather ponderous process.

BANGART: Unfortunately, historically, it has been the usual time span. Additionally, we have now to work with a new group within the NRC, the committee that reviews generic requirements. This replaced the Regulatory Requirement Review Committee that became known as the "Ratchet Committee." We have not worked with that group before so we don't know their ability to address these documents in a timely fashion. They were doing many, many things that are underway within the NRC.

FIRST: Are you suggesting that it is a more ponderous procedure now than it was prior to TMI?

BANGART: I am not sure the outcome will be a greater time frame. That may be one of the results.

CHRISTIAN: If I recall correctly, untreated filters can only claim 99% efficiency when taking credit. Is that correct?

BANGART: Yes.

CHRISTIAN: If you were to go back to requiring testing, would you permit 99.7% credit to be taken for installed filters?

BANGART: We have not addressed that point. The reason for only 99% credit is not based on the fact that DOE filter testing is not longer required. It is a conservative value that was chosen to use for our licensing guidance. Dr. Bellamy, who was a principal author of the current revision, may be able to address that point in greater detail.

BELLAMY: Mr. Bangart is right. The credit that we would allow a bank of HEPA filters after an accident is not correlatable to whether the filters were sent to a filter test station.

KOVACH, J.L.: What is the formal NRC position where the technical specifications of a plant refer to ANSI N510 (1975) through Reg. Guide 1.52, Rev. 2, even though there are mathematical errors in N510?

BANGART: The most correct action for a power plant to take would be to request an amendment to the technical specifications of its license to alter the wording of the testing requirement to remove the reference to the section of the standard which contains the errors. When asked by individual utilities and filter testing

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companies, however, our position has been that utilities, testing companies, and the NRC representatives performing inspections should consider N510-1980 as essentially equivalent to N510-1975 from a regulatory standpoint, since it can be successfully argued that N510-1980 is merely a more up-to-date and technically correct version of the earlier standard referenced by the nuclear power plants' technical specifications. The NRC will make a greater effort to inform industry of its current position in this matter.

SGALAMBRO: Could you give us some highlights on which items are going to be changed or revised in Reg. Guides 1.140 and 1.143?

BANGART: Revisions that are made in Regulatory Guide 1.52 will for the most part also be included in Regulatory Guide 1.140. The present practice by which Regulatory Guide 1.140 contains much of the same guidance as Regulatroy Guide 1.52, except for Engineered Safety Feature requirements, such as redundancy, seismic, and environmental qualification, will continue.

One of the major changes anticipated for Regulatory Guide 1.143 will be to add design guidance for radioactive waste incinerators that will be used in nuclear power plants to reduce the volumes of waste that are generated during plant operation. Within the last year we have also been challenged on the Reg. Guide 1.143 guidance which calls for Operating Basis Earthquake Seismic Qualification for foundations for those buildings which contain liquid radwaste management systems. We will be evaluating whether it is appropriate to continue with our current position on building foundation seismic qualification.

HOLLOMAN: As I understand from previous conversations, QPL qualification is not currently required for HEPA filters by NRC. Will this be considered for possible inclusion in the revision of Reg. Guide 1.52?

BANGART: NRC will probably remain divorced from Mil 51068-QPL, but this point will be evaluated in the process of revising Regulatory Guide 1.52. Other industry representatives who feel that QPL qualification should be addressed in Reg. Guide 1.52 are invited to comment to the NRC.

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IN-PLACE HEPA FILTER AEROSOL TEST SYSTEM

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Abstract

An In-Place HEPA Filter Aerosol Test System that exhibits the characteristics of a low cost, reliable, safe method of testing the integrity of individual filter stages (banks) of multiple-stage, high efficiency particulate air filter (HEPA) systems has been developed. The In-Place HEPA Filter Test System (HFTS) eliminates problems of conventional in-place testing and conforms with the intent of ANSI/ASME N510-1980 as it applies to testing HEPA filter systems in nuclear air cleaning systems.

Rockwell Hanford Operations (Rockwell) at Richland, Washington designed, fabricated and in June 1980, successfully tested the In-Place HEPA Filter Test System. The tests of the HFTS demonstrated the ability of the system to perform the functions required to evaluate the integrity (leak tightness) of individual stages for a multistage HEPA filter installation. The system is especially applicable to HEPA filter systems that are physically inaccessible or highly radioactive, where contact maintenance cannot be performed as required by other in-place type test systems.

I. Introduction

An In-Place HEPA Filter Aerosol Test System (HFTS) was designed and a prototype fabricated and tested by Rockwell Hanford Operations (Rockwell) of Richland, Washington. The prototype of the HFTS was successfully tested in June 1980. The test demonstrated the ability of the HFTS to perform the functions required to evaluate the integrity (leak tightness) of individual stages for a multistage (bank) HEPA filter installation. The test also demonstrated that the intent of ANSI/ASME N510-1980 (ANSI N510-1980) was being met. The test was performed using dioctyl phthalate (DOP) as the challenging trace agent; however any aerosol suitable for testing HEPA filters can be used.

Background and Problems

In-place testing of HEPA filter systems is necessary to verify the high performance levels required of a nuclear air cleaning system. The usual test for in-place testing, in the United States, for high efficiency filter systems used in nuclear application is a dioctyl phthalate (DOP) penetration test performed in accordance with ANSI N510-1980. In this method a smoke cloud of poly-dispersed DOP is generated by a DOP aerosol generator and dispersed uniformly over the upstream face area of the filter stage being tested. Any smoke which penetrates the body of the filter or leaks through gasket or filter frame cracks, etc., passes into the downstream region where it is thoroughly mixed with the filtered air. This concentration is then compared to the upstream concentration and the difference is expressed in percent penetration.

The nuclear air cleaning system usually contains two or more HEPA filter stages in a series in the same containment. This arrangement and the laminar flow that exists between the stages creates special problems to in-place testing. The problems are the difficulty of obtaining an adequate mixing for a single point representative penetration sample and uniform distribution of trace agent concentrations between the stages. Space limitations and cost considerations usually dictate the design arrangement of multistage filters to the extent that the banks are so close together that neither of these objectives can be achieved. This difficulty is further complicated in a system that is subjected to a hostile environment where human occupancy is not permitted or the system is physically inaccessible. The very high collection efficiency of the first-stage filter prevents sufficient trace agent from being introduced upstream of the second stage to permit effective testing of the second stage. (Fig. 1) Adequate mixing of clean air and trace agent penetration can usually be accomplished downstream of the last filter stage for a representative sample, but the very high collection efficiency of the second stage filter does not allow sufficient trace agent to penetrate this barrier to permit a representative sample from the first stage. (Fig. 2)

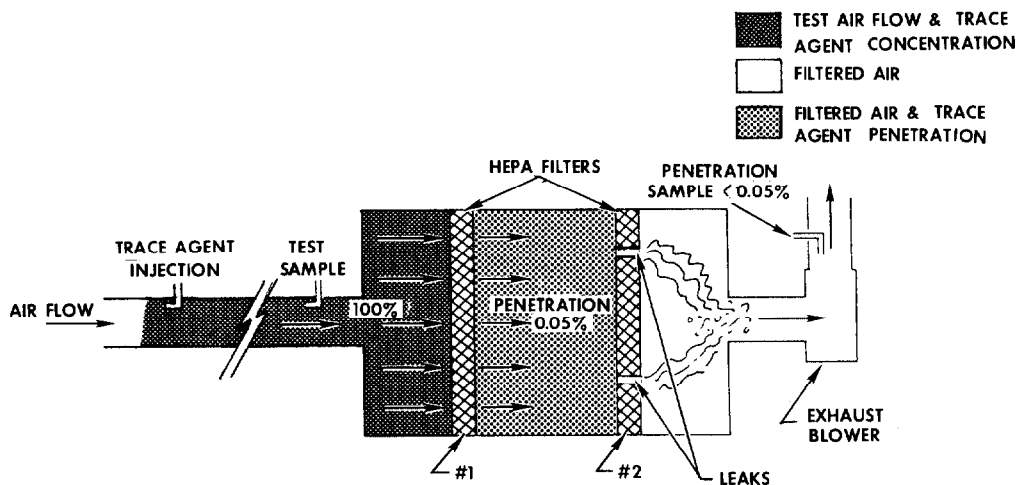


FIGURE 1. LEAKS IN SECOND STAGE NOT DETECTED

Prior Test Procedures

In-place testing of individual stages for single and multistage HEPA filter installations, to be in compliance with ANSI N510-1980, consists of the use of the installed duct work, temporary injection and bypass ducts, moveable orifices and mechanical and electrical devices (fans) and contact maintenance (probe-scanning). These are necessary to achieve adequate dispersion, mixing and sampling of the aerosol test agent. The disadvantage to probe-scanning, temporary duct work and moveable mechanical and electrical devices is the requirement for contact maintenance. Contact maintenance results in radiation exposure to personnel and in some nuclear air cleaning systems, radiation is of such magnitude that personnel access is prohibited.

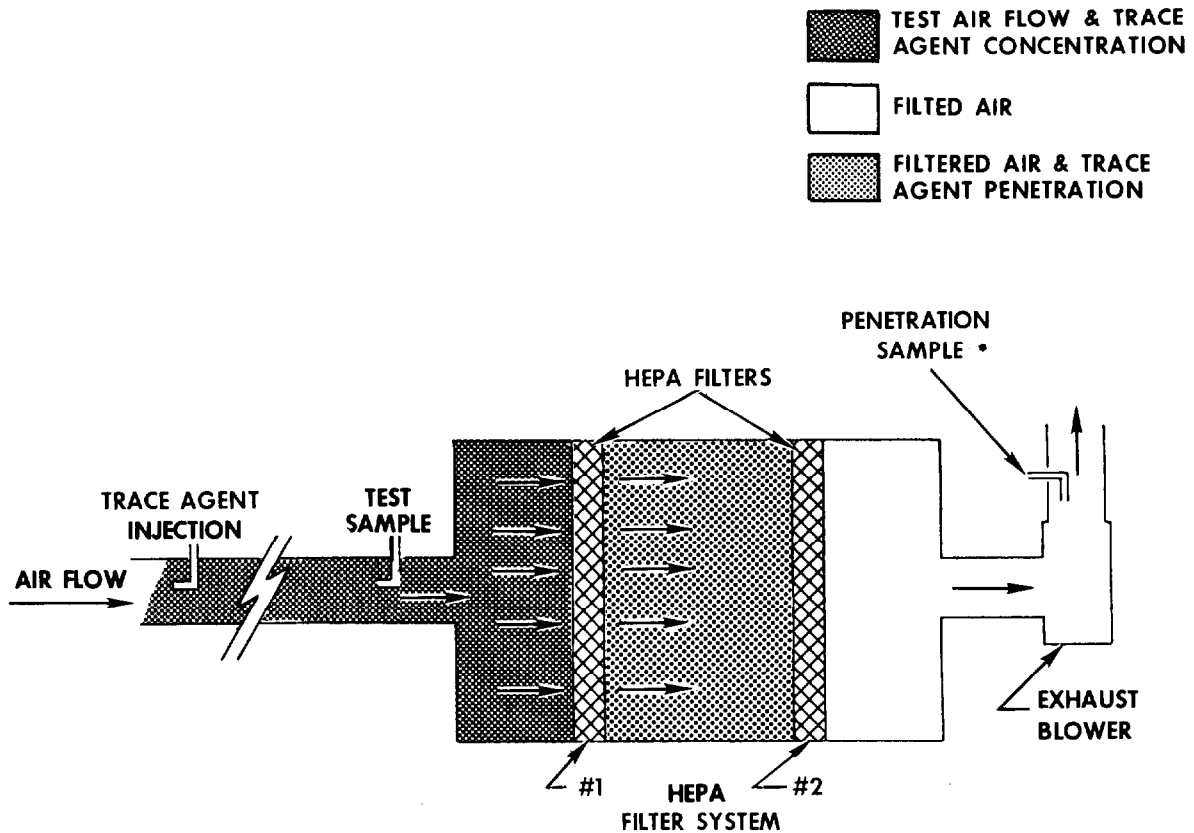


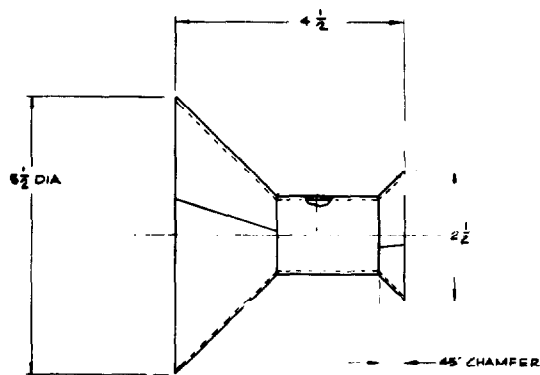
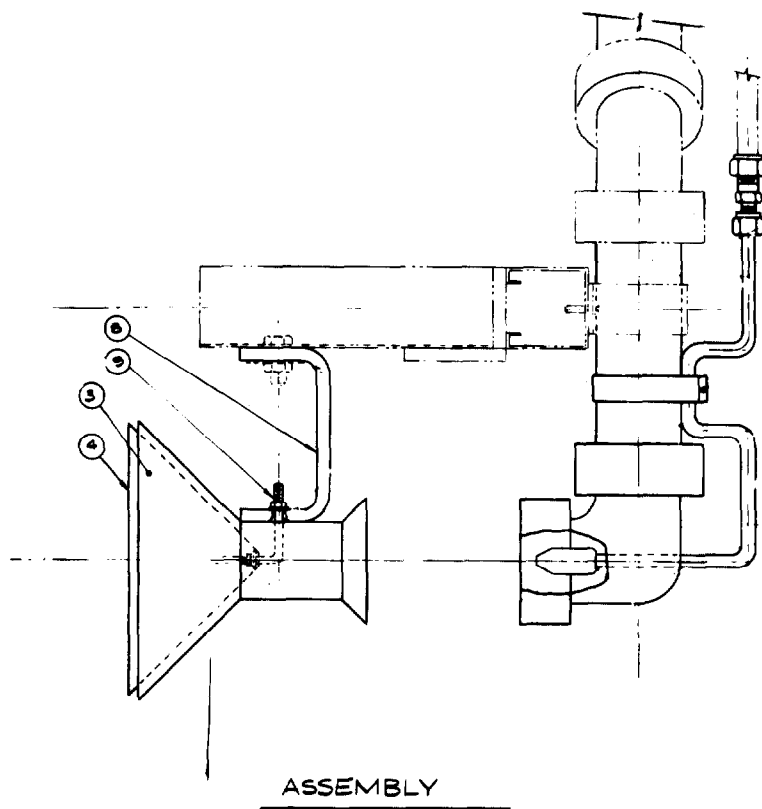
FIGURE 2. PENETRATION SAMPLE IS NOT REPRESENTATIVE OF FILTER BANK #1 PENETRATION

II. Description and Operation

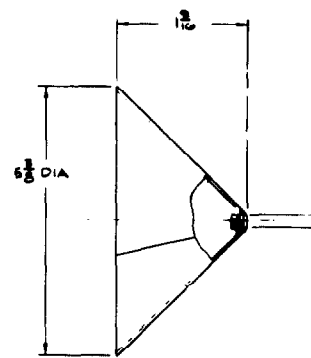
The Rockwell In-Place HFTS consists of Diffuser/Ejector (D/E) (Fig. 3) assemblies and single point sample ports (SP) (Figs. 4a and 4b), which are located upstream and downstream of the filter stage under test. In most arrangements, the D/E's and SP's are located between each filter stage for which testing is desired. (Fig. 5) This provides permanent trace agent injector and sample ports.

The D/E assembly performs two functions:

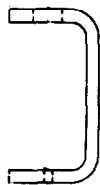
1. As a diffuser the device creates turbulence in the laminar flow between the filter stages to insure good mixing of the clean air and trace agent that has penetrated the filter stage under test. Adequate mixing is necessary to obtain a representative sample at a single point. (Fig. 6a & 6b)
2. As an ejector the device introduces the trace agent into this turbulent flow to provide a uniform trace agent dispersion over the upstream face area of the stage being tested. (Fig. 7)



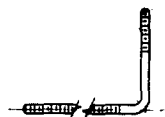
(3) ASSEMBLY



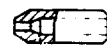
(4) ASSEMBLY



(8)



(9)



(7)

FIGURE 3. DIFFUSER/EJECTOR

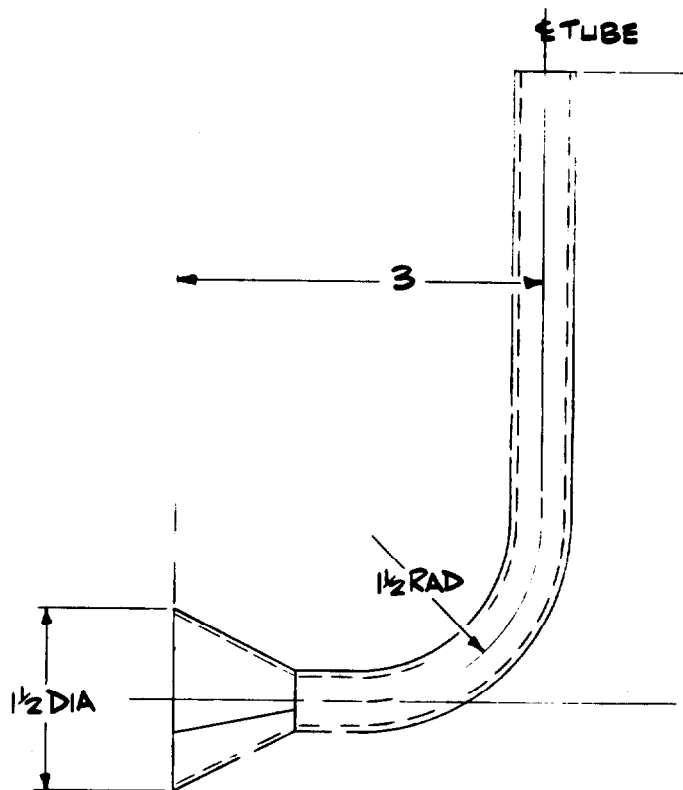


FIGURE 4a. SAMPLE PORT

The D/E device can be constructed of metal or plastic components. The device consists of an inner and outer cone, a high velocity pressure chamber, a high pressure nozzle and associated piping and tubing, as shown in Fig. 3 and 8.

The D/E device is designed to create turbulence in a laminar air flow with a minimum resistance to the flow. This characteristic is desirable to disperse the trace agent uniformly over the face area of the filter stage being tested, as shown in Fig. 7, and to agitate the clean air trace agent concentration downstream of this stage, so that a representative sample of the penetrating trace agent can be obtained at a single sample port as shown in Fig. 6a.

The principle involved in the operation of the device is fluid flowing at high velocity creates a low pressure. This principle is applied at several integral operations without mechanical or electrical manipulation. When operating as a diffuser, it generates turbulence in a laminar flow. (Fig. 6a) This is accomplished by a jet venturi action, where a high pressure air flow creates a low pressure and part of the laminar flow is drawn in by the low pressure into a high pressure chamber. The release of this pressure is by flow through an annular space between two cones, again, creating a low pressure disturbance to the remaining laminar flow. This action creates an agitated homogeneous mixture in a normally laminar flow condition. As an ejector, the device operates the same as a diffuser, except the trace agent is drawn into the low pressure area, compressed and dispersed through the device and ejected into the turbulent air flow, again creating a homogeneous mixture as shown in

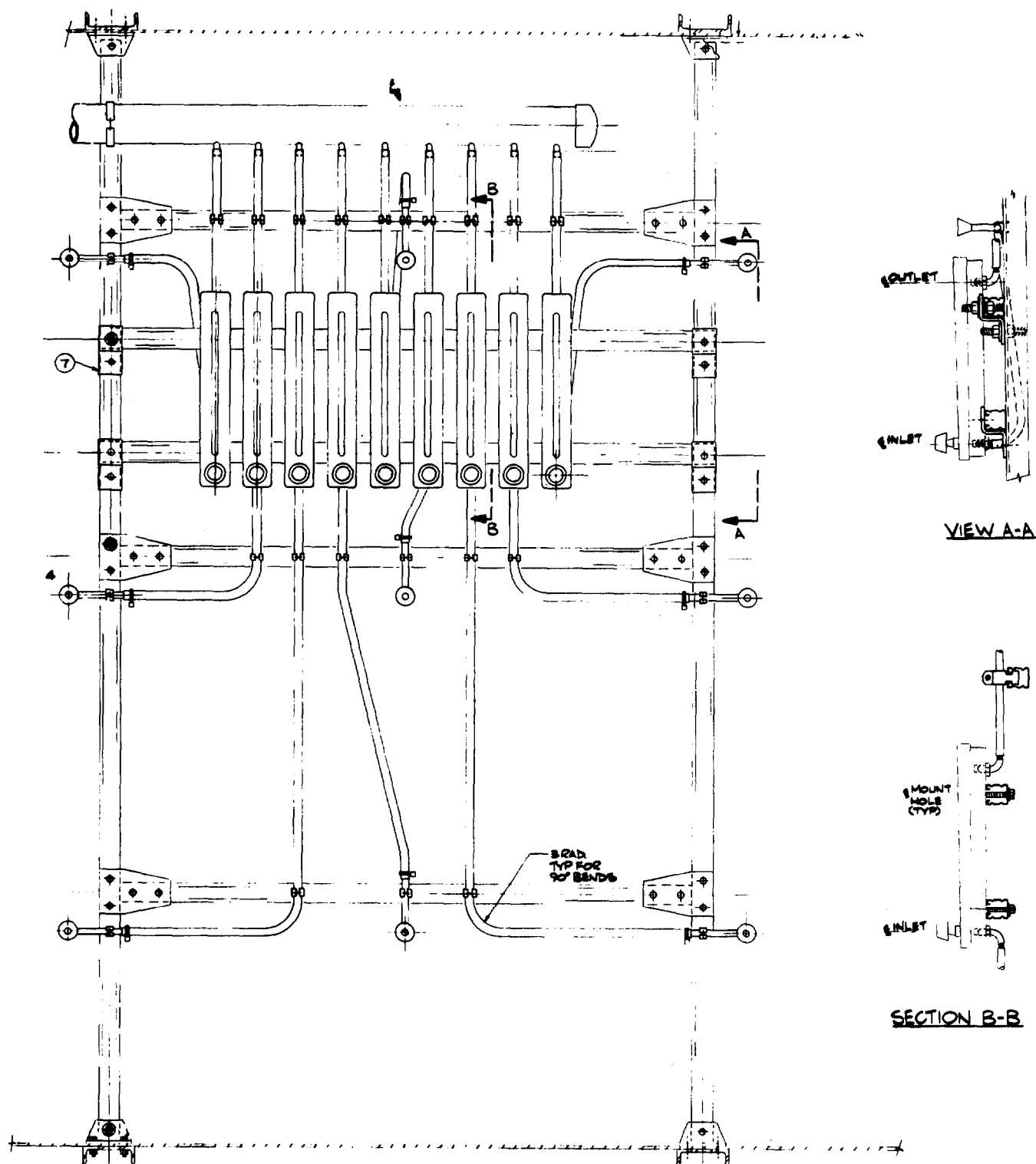


FIGURE 4b. NINE PORT SAMPLE PORT ARRAY

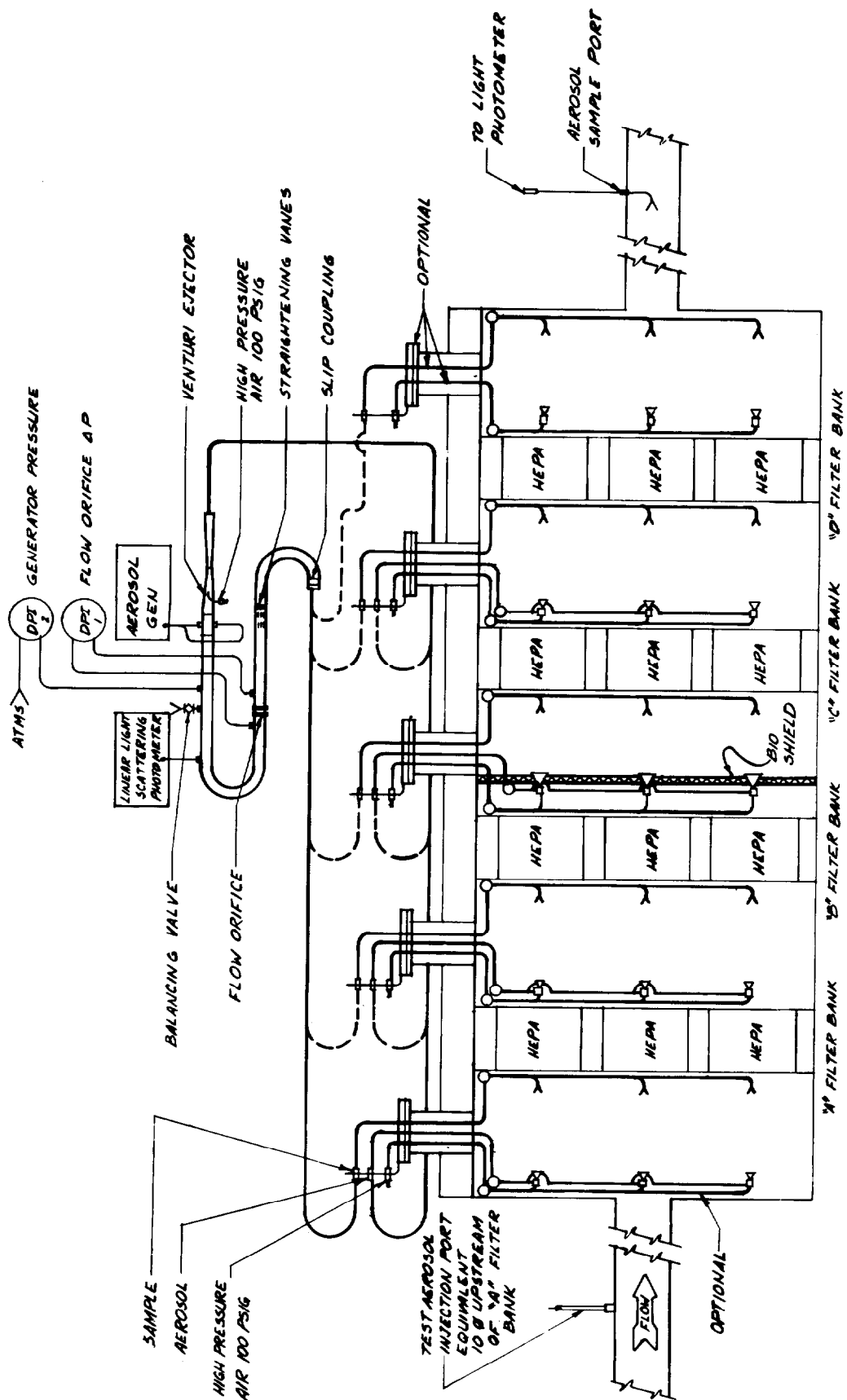


FIGURE 5. DIAGRAM OF A ROCKWELL IN-PLACE HFTS

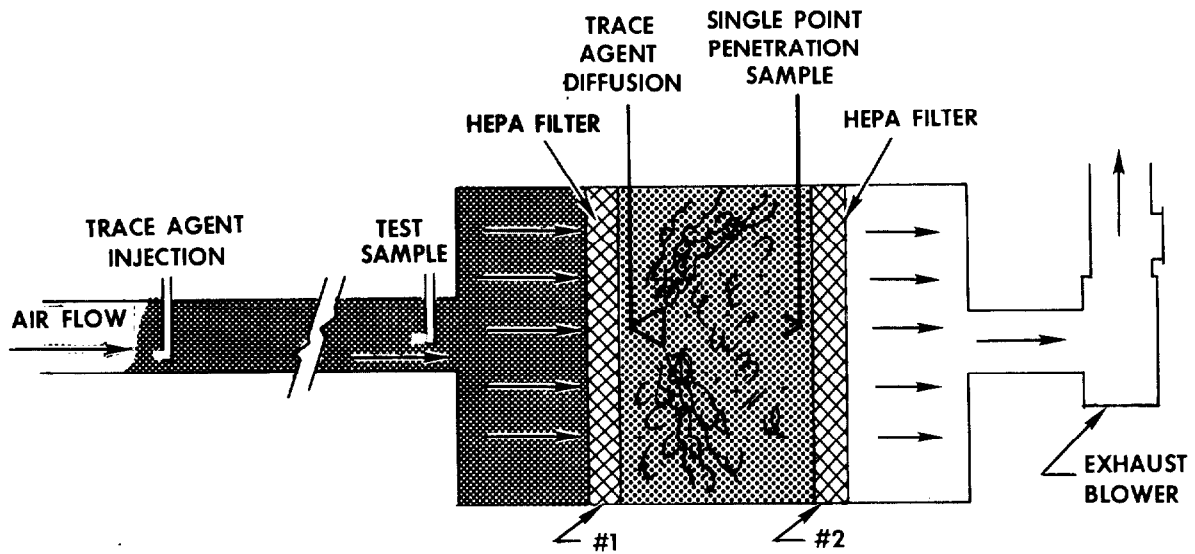


FIGURE 6a. FIRST STAGE PENETRATION TEST OF TWO-STAGE HEPA FILTER WITH HFTS AGITATION

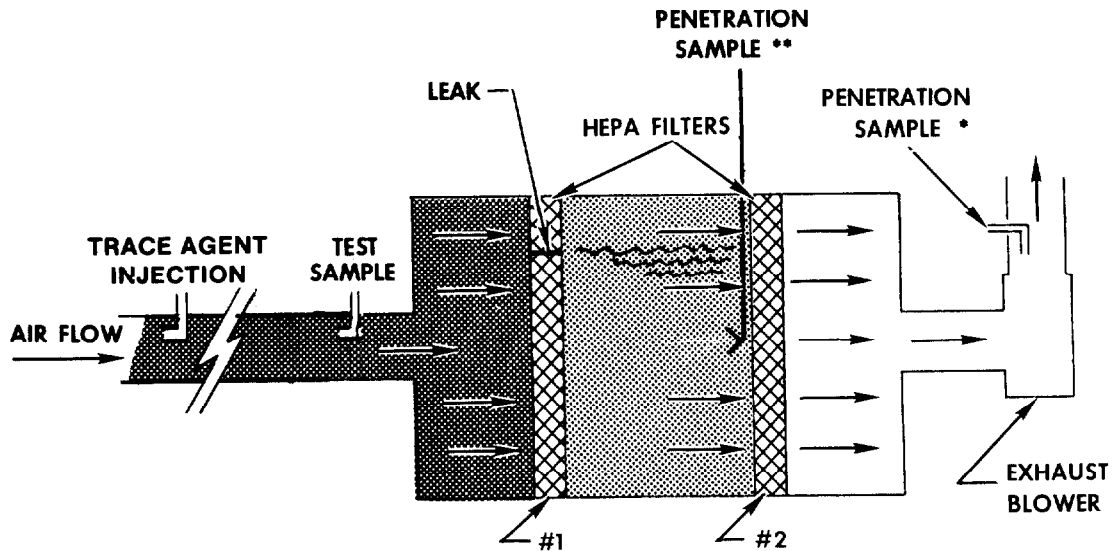


FIGURE 6b. FIRST STAGE PENETRATION TEST WITHOUT HFTS AGITATION

*Penetration sample is not representative of filter bank #1 penetration.

**Without agitation to the laminar flow the sample of the leak is not detected by the single point.

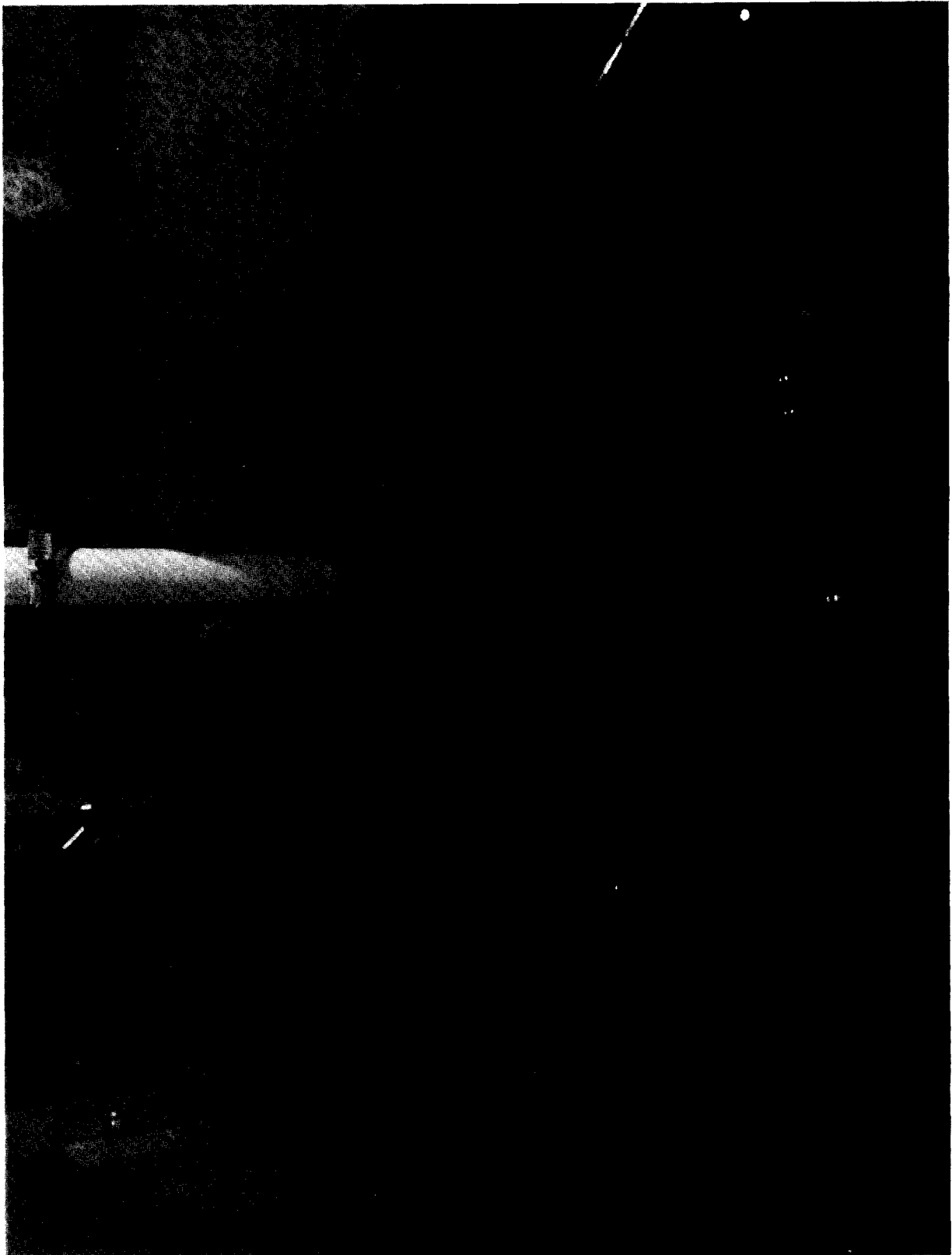


FIGURE 9
Diffuser/Ejector Device operating as an ejector
dispersing DOP Test Agent.

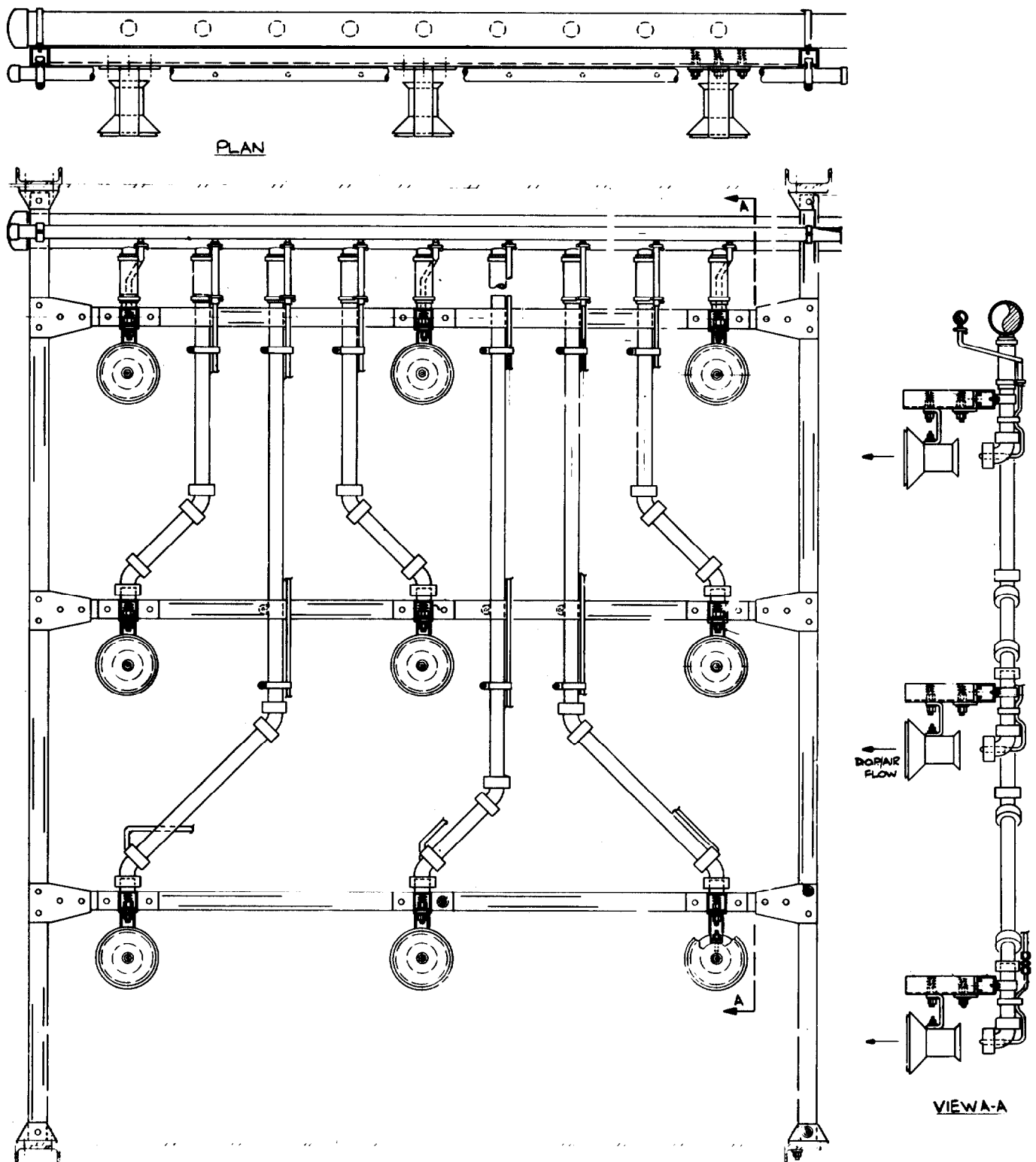


FIGURE 8. NINE DIFFUSER/EJECTOR ARRAY

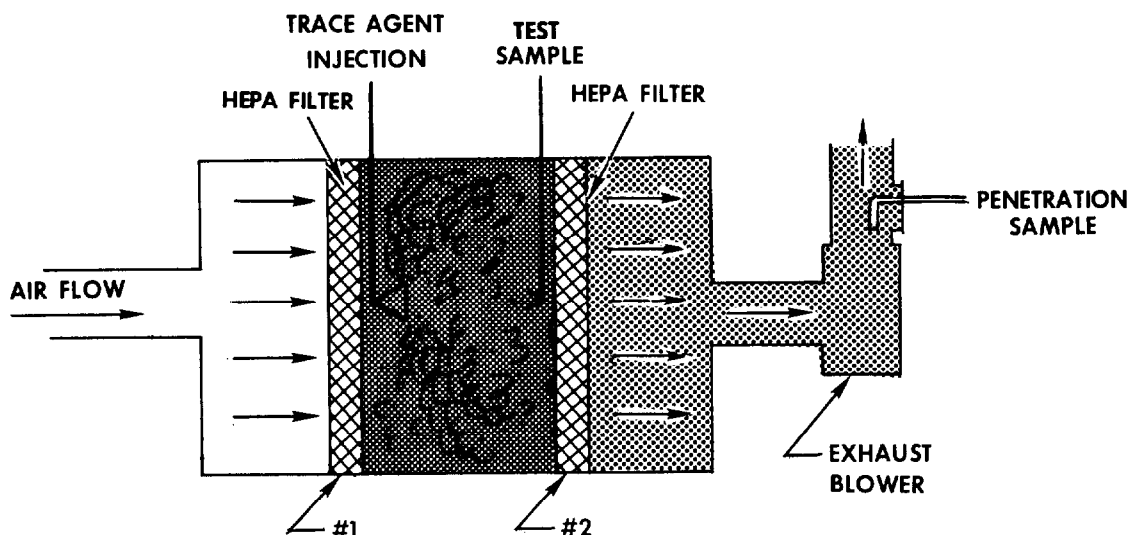


FIGURE 7. SECOND-STAGE PENETRATION TEST OF TWO STAGE HEPA FILTER WITH ROCKWELL IN-PLACE HFTS

Fig. 7. In this manner the device mixes and ejects the trace agent into the flow so a uniform dispersal can be achieved. This action is shown in Fig. 9; the test agent, DOP, is being injected into a laminar air flow with agitation. Figure 10 shows the test agent, DOP, being induced into the laminar flow without agitation. As a diffuser, the device mixes the penetrant trace agent with the flow in the same manner so a representative sample can be obtained at a single sample point.

III. Testing

Test Introduction and Purpose

The purpose of performing an in-place test on the laminar flow multiple bank (HEPA) filter installations is to determine the integrity (leak tightness) of the individual filter banks (filter, gasket seals and filter frames) as an assembly.

The purpose of the test procedure was to provide a means of evaluating the Rockwell in-place HFTS for compliance with the intent of ANSI N510-1980.

The goal of conducting the test for the Rockwell in-place HFTS was to verify the capability of the system to provide a means of determining the integrity of the individual filter stages (banks) with reliable repeatability. This requires a system that will obtain representative samples of the trace agent penetration and trace agent from 0.001% to 100% and uniformly dispersed trace agent within $\pm 10\%$ over the upstream face area of the bank being tested.

The test filter box assembly that was used for the qualification and verification test housed two stages of HEPA filters. It was designed so actual HEPA filter installation characteristics could be duplicated for test purposes. A flow tube and control



FIGURE 10
DOP Trace Agent being induced into a laminar flow without agitation.

damper were attached to the intake upstream transition and a connecting duct, control damper and exhaust fan were connected to the (exhaust) downstream transition. The arrangement of the flow tube, filters, D/E assembly, sample port, downstream duct and exhaust fan are shown in Figs. 11, 13 and 14.

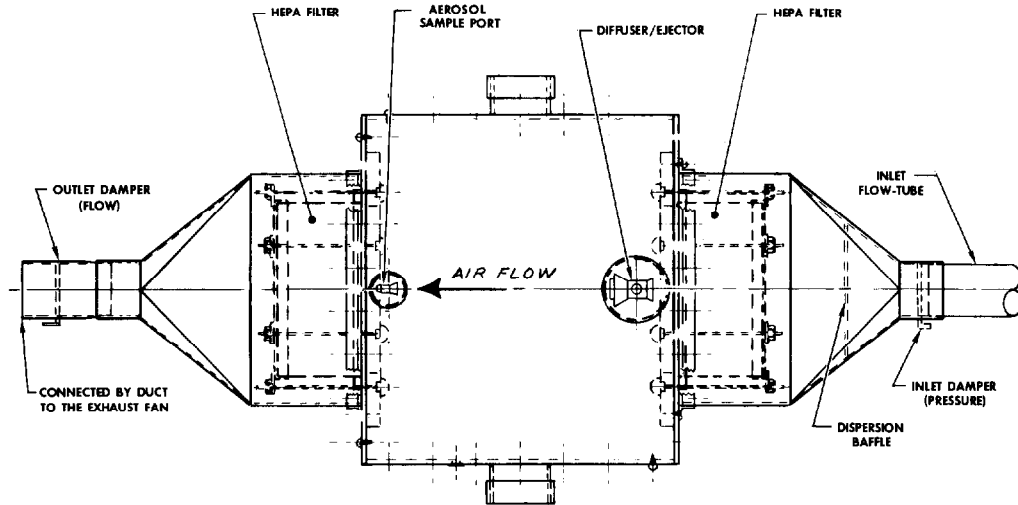


FIGURE 11. TEST FILTER ASSEMBLY

Support components include a source of compressed air, aerosol generator, photometer (particle detection apparatus) and an aerosol flow orifice injection assembly. The aerosol flow orifice injection assembly provides a simple means of supplying the test aerosol to the diffusers and extracting the test sample. (Fig. 12)

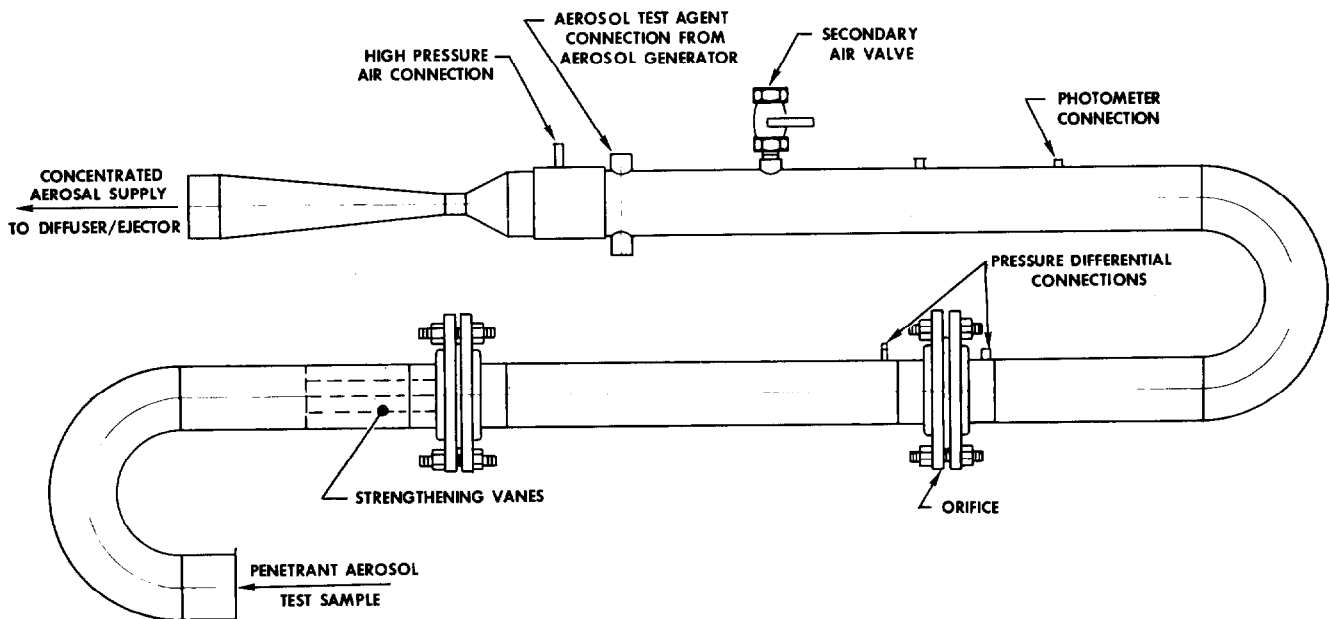


FIGURE 12. FLOW ORIFICE INJECTION ASSEMBLY



FIGURE 13. FLOW TUBE AND CONTROL DAMPER

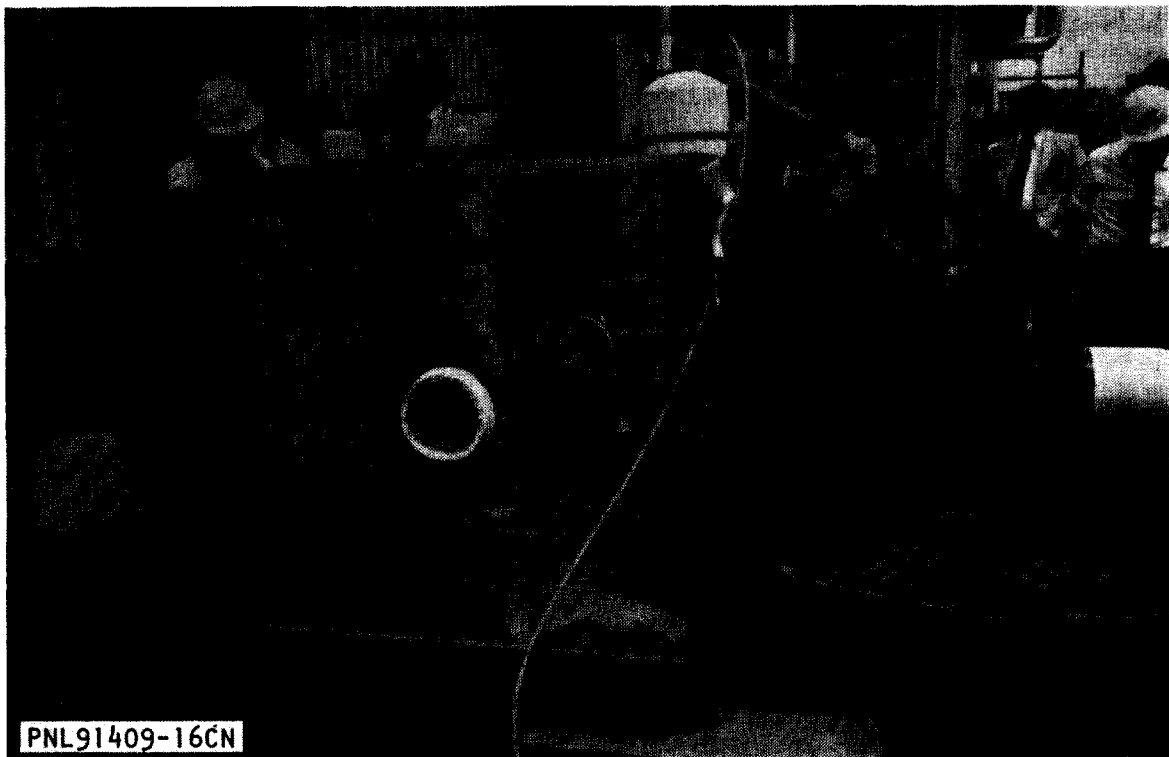


FIGURE 14. TEST FILTER, TRANSITION AND HOLDER ASSEMBLY

Test Run

The first step of the test procedure established an environment similar to an actual operational filter system. The air flow was adjusted to 1250 CFM and a comparison of the filter performance was made to the Department of Energy test station data.

The next step verified that external contaminants, such as box leakage, dust or detectable gases entering the downstream area of the first filter, were not present to influence the outcome of the test. The D/E high pressure air did not inject dirt or oil into the air stream. No adverse conditions were detected.

The next step demonstrated that a representative sample taken anywhere over the upstream face area of the second filter could detect any credible leak located anywhere on the first stage (bank). This was possible only when the D/E was in operations. This step was repeated several times with the leak in various locations. Repeatability in detecting the leak was excellent.

The next step was removing the downstream filter and comparing the in-place HFTS results with those of conventional in-place test results. This portion of the test was also performed several times with the same results. The single point sample taken with the D/E in operation was equal to the sample taken 10 duct diameters downstream of the filter under test.

The next step of the test procedure was to demonstrate the capability of the D/E to disperse DOP aerosol uniformly over the face area of the test filter at the design air flow of 1250 CFM. For this portion of the test 100% DOP aerosol concentration was dispersed over the face area of the test filter; uniformity was within 10%. This step in the test procedure was repeated several times with excellent results. The uniformity of the DOP aerosol dispersion over the face area of the filter under test was within 5% to 10% of saturation. In the subsequent tests, further comparison showed that conventional in-place test results were not as good (less uniformity) as those of the Rockwell In-Place HFTS.

Further testing determined the consequences for the possibility of moisture accumulations in the high pressure air lines which could take place in an actual installation. The D/E, acting as an atomizer, dispersed the water into droplets so small that there was no damage to the filter media of the HEPA filter.

The results of the prototype test demonstrate that in-place tests performed with the Diffuser/Ejector device are superior to those performed in the conventional procedures and are in conformance with the intent of ANSI N510-1980. The cloud of DOP smoke as shown in Fig. 9, is uniformly distributed over the upstream face area of the filter stage under test and when the D/E is operated as a diffuser, the downstream DOP penetration and clean air are thoroughly mixed so a representative sample can be obtained at a single sample point.

Test Equipment

The equipment used to test the In-Place HFTS was the same as that used by the HVAC test group for routine maintenance of HEPA filter installations throughout the Hanford area. Routinely inspected and calibrated, the equipment has current certification.

The particulate detection apparatus, manufactured by Air Techniques, Inc., is a self-contained forward light-scattering photometer, Model TDA-2DN. The photometer has a baseline response which may be set as a basis of comparison, using the internal calibration circuit, which is set for 100 micrograms of 0.3 micron particles. The user can also set the baseline response against any concentration of matter up to 1×10^{20} particles/ft³ whose particle size may range from 0.2 micron up to 100 microns. Once the baseline has been set, any subsequent readings will be in relation to that baseline. The readable range of concentration is from 0.001 to 100.

The aerosol generators are of the thermal and compressed air types. Also manufactured by Air Techniques, Inc., the thermal unit, Model TDA-5A generator, vaporizes and reconstitutes the trace agent into a polydispersed aerosol. The compressed air generator is a Larkin unit with eight nozzles. The gas-thermal and compressed air generators produce a polydispersed aerosol of about the same NMD and size.

Dwyer Instruments, Inc. manufactures the magnehelic and flex-tube U-tube differential pressure gauges. Air flow is measured by a pitot tube, inclined manometer and a series 6000P velometer made by Alnor Instrument Company. Appendix D contains the test equipment specification sheets.

III. Conclusion

Conforming to the intent of ANSI N510-1980, this new design provides a means of testing the integrity (leak tightness) of the individual filter stages (banks) in a multistage filter installation without contact maintenance. The prototype of Rockwell's In-Place HFTS demonstrated the capability of the system not only to meet but surpass these requirements.

Future use of the Rockwell In-Place HFTS is unlimited in the field of testing HEPA filter systems because the feature of almost instantaneous dispersal and mixing of the test agent makes its application desirable to filter systems where contact maintenance can be performed, as well as for the system where contact maintenance cannot be performed. This could mean a tremendous savings in the operating cost of HEPA filter systems in the savings of manhours and trace agent material. Typical applications of the Rockwell In-Place HFTS are shown in Fig. 15 and 16.

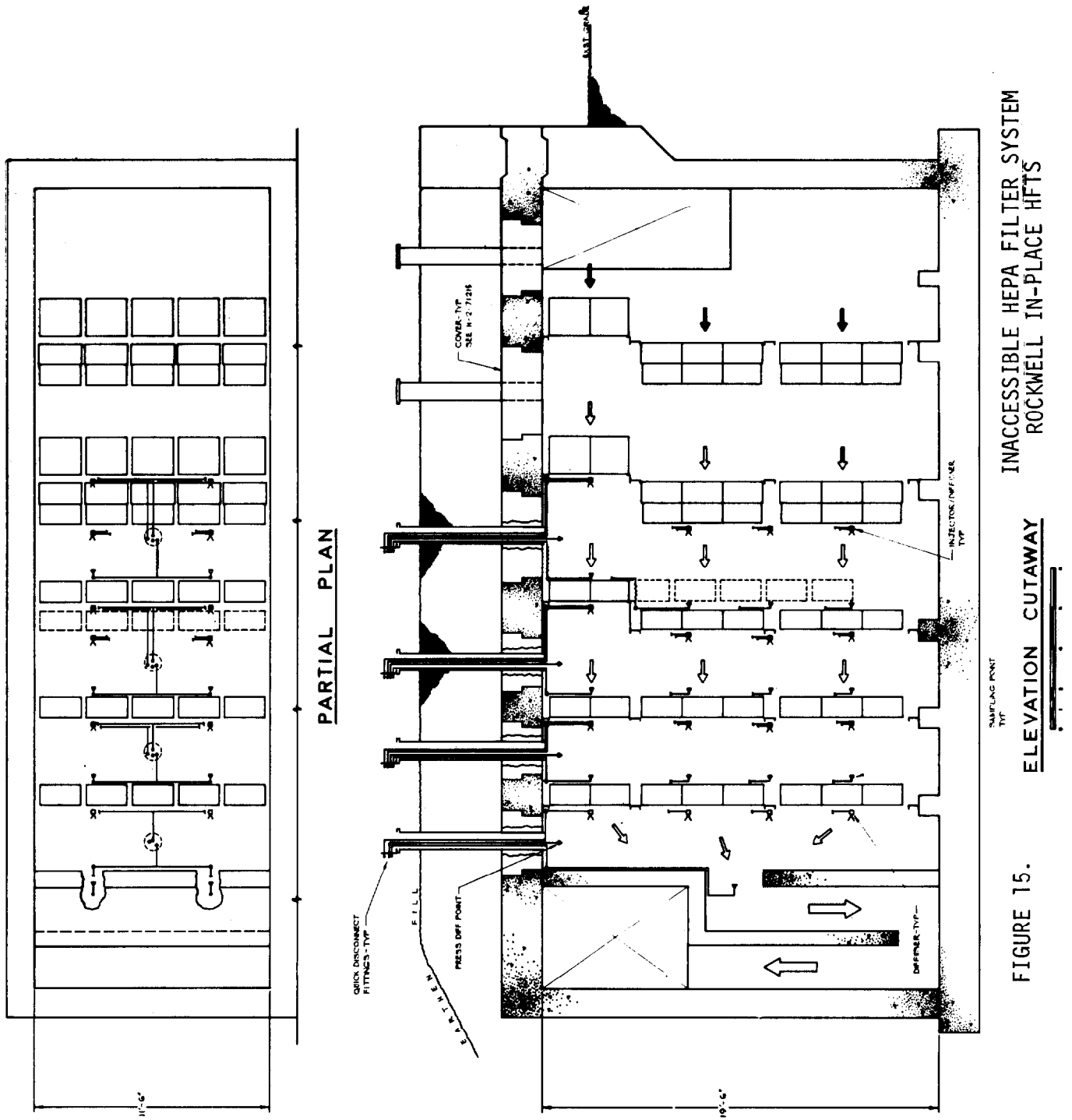


FIGURE 15.

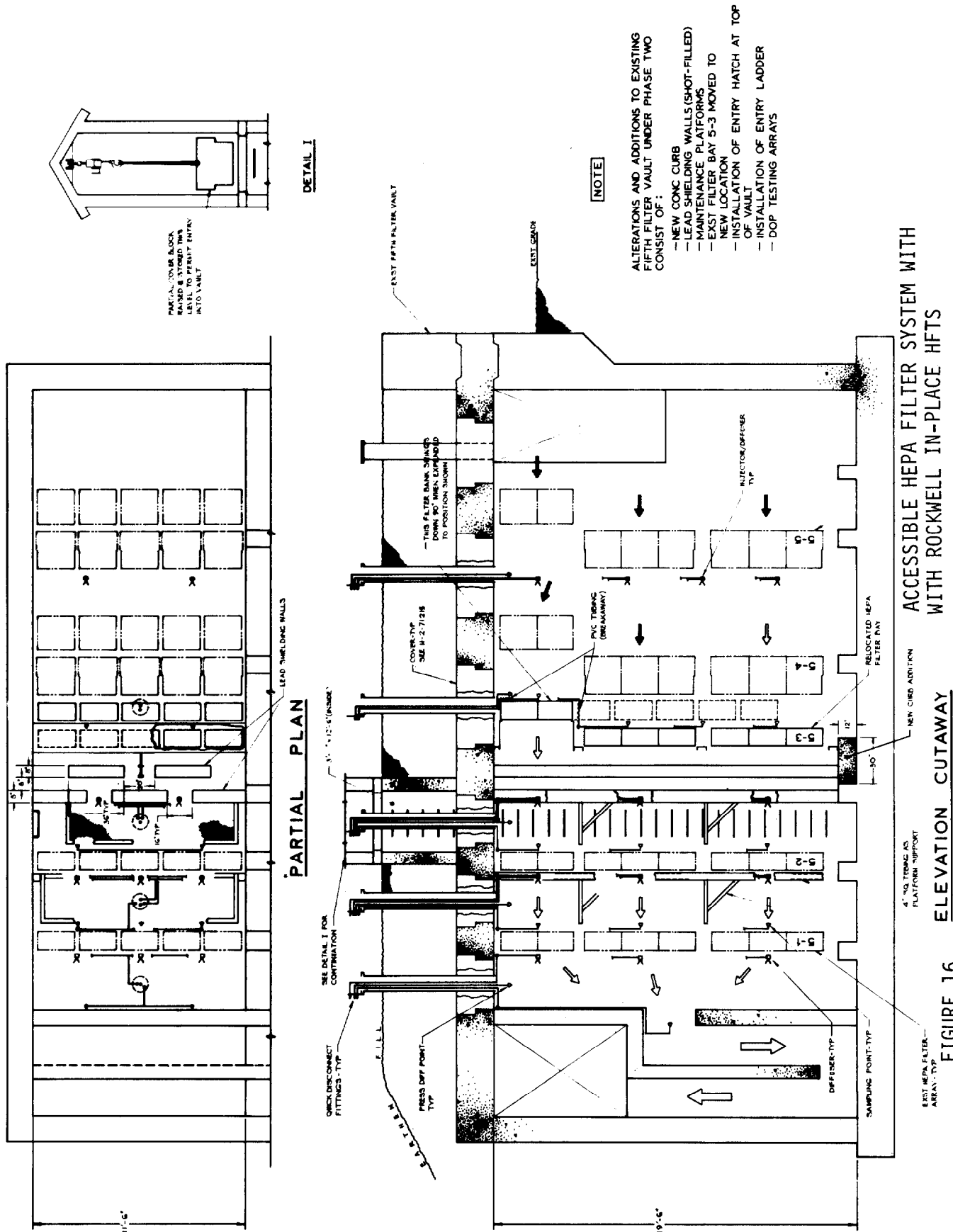


FIGURE 16.

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1. Laskin, S. "Submerged Aerosol Unit", AEC Project Quarterly Report-UR-38, University of Rochester (1948)
2. Burchsted, C. A., "Nuclear Air-Cleaning Handbook", ERDA 76-21 Oak Ridge National Laboratory, Oak Ridge, Tennessee, Second Edition 1976
3. ANSI-ASME N510-1980, "Testing of Nuclear Air Cleaning Systems", The American Society of Mechanical Engineers
4. First, N. W. & Gilbert, H., "Aerosol Filtration", Harvard School of Public Health, 16th DOE Nuclear Air Cleaning Conference, Volume I, Page 633 through 663. References included therein are applicable to this document.

DISCUSSION

DUPOUX: In many filter casings, in France especially, the distance between the two HEPA filters in tandem is very short and I wonder if your experiment would apply very often? Have you determined the minimum value of the ratio defined by the distance between the filters divided by the size of the filters, still permitting correct sampling to test the upstream filter and an homogeneous concentration upstream of the downstream filters?

HERMAN: The device can be placed anywhere. For a 24" x 24" filter, the minimum distance is about 8 inches. That would be about as close as you can get to it. If you want to cover a 2 x 2 bank of filters, i.e., 4 filters, you would be back about three feet if you wanted one diffuser to cover all four filters.

PRATT: A couple of points, what volume of compressed air is injected and are the dimensions of the test rig in terms of the area of the transition between filters and downstream duct diameter typical of plant sizes?

HERMAN: The compressed air requirement is about 1.5 CFM. With regard to dimensions, we tested it at that particular distance and we also tested it just 12 inches away. We simply blanked off a square area the size of the filter and came up within 12 inches of it. This was done to show that you could get adequate dispersion over a very large surface if you had at least four feet of distance. You could almost cover a three by three filter bank, i.e., 9 filters, with one diffuser at about four feet distance.

HOLLOMAN: 1- Is the same device used to mix the aerosol downstream of the filter as is used to inject DOP upstream? If not, how does the diffuser/ejector differ from the diffuser downstream of the filter prior to sampling? What is the distance from filter to downstream mixing and sample points? 2- How was the upstream DOP dispersion measured? 3- How many sample ports for a 24" x 24" filter?

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HERMAN: 1- The same device is used for dispersing the test agent as is used for mixing the penetrant and clean air. Depending on the number of units for the area to be covered, 12 to 36 inch spacing is usually adequate. 2- By traversing the face area with a sample probe. 3- One sample port.

WATSON: What is the minimum distance of a sampling cone from the downstream face of the test filter needed to get a representative sample from the area sample by that cone?

HERMAN: Minimum distance between stages for a 24" x 24" HEPA filter would be 12". (Approximately 8" between the diffuser/ejector and the sample port.)

FURRER: You measured different efficiencies as a function of time. Is that a function of a changing aerosol size distribution during your process?

HERMAN: Particulate size remained about the same when compared to a conventional dispersion, i.e., DOP injection 10 duct diameters upstream of the sample plane.

OLDHAM: Is uniformity of mixing achieved when the transition stage is the same size as the filter stage? If not, do you have to oversize the transition stage to achieve the necessary turbulence to overcome laminar flow in any size system? How was uniformity determined?

HERMAN: Adequate test aerosol dispersion and penetrant sample mixing can be achieved for a 2' x 2' area at approximately 12" spacing with one diffuser ejector and one sample port. A 4' x 4' area can also be covered with one unit with 36" between stages. Uniformity was determined by traversing the test area with a sample probe.

FISH: In large banks, multiple diffusers would be used. Are multiple samplers used? The sample lines may be of extended length. Was any check made to see if DOP plate-out takes place in the sample lines?

HERMAN: The distance between the banks determines the number of diffuser/ejectors and the sample ports. Each 24" x 24" filter requires a diffuser/ejector and sample port at 12 inch spacing. DOP plate-out does not seem to be a problem.

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IN-PLACE REALTIME HEPA FILTER TEST METHOD*

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ABSTRACT

High Efficiency Particulate Air (HEPA) filters are essential in all phases of the nuclear industry. Their installation is followed by in-place efficiency testing and subsequent periodic retesting before a filter is replaced. However, if each filter's efficiency could be continuously monitored, breached filters could be immediately identified, thereby improving the testing and replacement process.

Previously reported work^(1,2) demonstrated the ability of an electrofluidized bed to continuously collect radioactive submicron particles. The present work reports the adaptation of this technique to the continuous in-place measurement of HEPA filter collection efficiencies.

Two heated electrofluidized beds, one upstream and one downstream of the filter, continuously sample the challenging and penetrating aerosol. The aerosol's activity is continuously monitored using two NaI (Tl) detectors. The ratio of the change in the two activities is proportional to the in-place filter efficiency.

The significant conclusions drawn from in-plant testing of this monitor were that the monitor could accurately measure a HEPA filter efficiency >99% under actual process off-gas conditions, could recover satisfactorily from condensation in the off-gas line, and it could be made insensitive to volatile forms of ^{106}Ru .

INTRODUCTION

The purpose of this work was to develop and demonstrate an on-line, realtime HEPA filter efficiency monitor. The selected approach was to measure the challenging and penetrating aerosol in situ with an electrofluidized bed.^(1,2)

A fluidized bed is a column of granular material that has sufficient gas flow directed through the column that the bed particles behave as a fluid and assume the shape of the containing vessel. An electrofluidized bed (EFB) has a potential gradient applied across the bed to increase particulate retention through electrostatic attraction. A diagram of a typical electrofluidized bed is shown in Figure 1.

Previous work^(1,2) with EFB's has established the following:

- 1) Collection efficiencies >99% can be achieved for particles down to $0.002\text{ }\mu\text{m}$ with a 30 cm bed operating at potential gradients $>3.5 \times 10^5\text{ Vm}^{-1}$;

* Work performed under US DOE contract DE-AC07-791D01675.

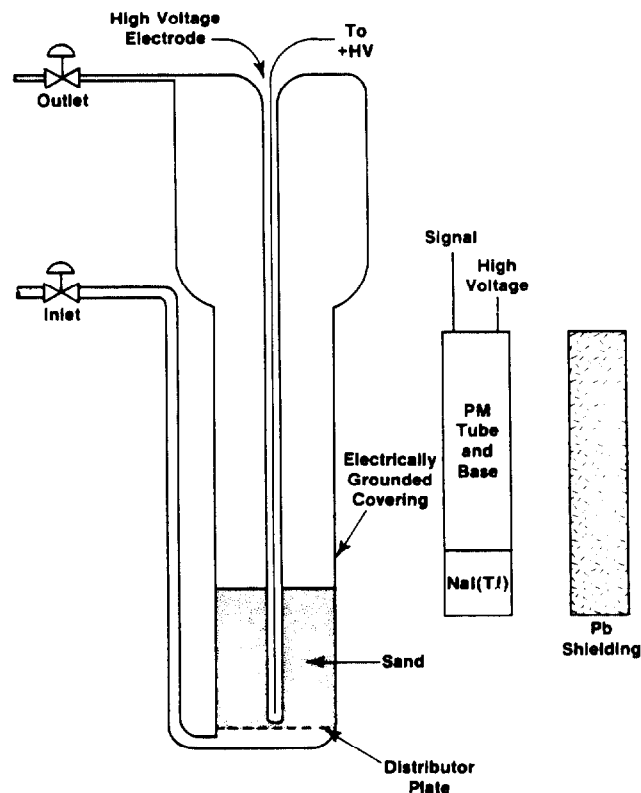


FIGURE 1 ELECTROFLUIDIZED BED WITH DETECTOR

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2) Collection efficiency increases with increasing temperatures;

3) Collection efficiency is not dependent on particle size for the range 0.002 to 0.200 μm .

Accordingly, this previous work indicated that an EFB was an appropriate device for collecting the radioactive particles that penetrate a HEPA filter. Once collected in this fashion, either the gross gamma activity or a single gamma emitter, depending on the application, could be monitored. The concept is shown schematically in Figure 2.

To calculate the HEPA filter efficiency for a system such as that of Figure 2, the following equations are used:

$$C = \frac{K \left(\frac{dA}{dt} \right)}{V} \quad (1)$$

where: C is the concentration of the off-gas;
K is the efficiency of the detector;

$\frac{dA}{dt}$ is the change in count rate with respect to time;
 V is the volumetric flowrate;

$$\text{and Penetration} = \frac{C_d}{C_u} \quad (2)$$

where: C_d is the downstream concentration (from Equation 1)
 C_u is the upstream concentration (from Equation 1)

$$\text{and } E = (1 - \text{Penetration}) \times 100\% \quad (3)$$

where: E is the HEPA filter collection efficiency in percent.

The testing of this concept was performed in two phases: a laboratory evaluation and an in-plant demonstration.

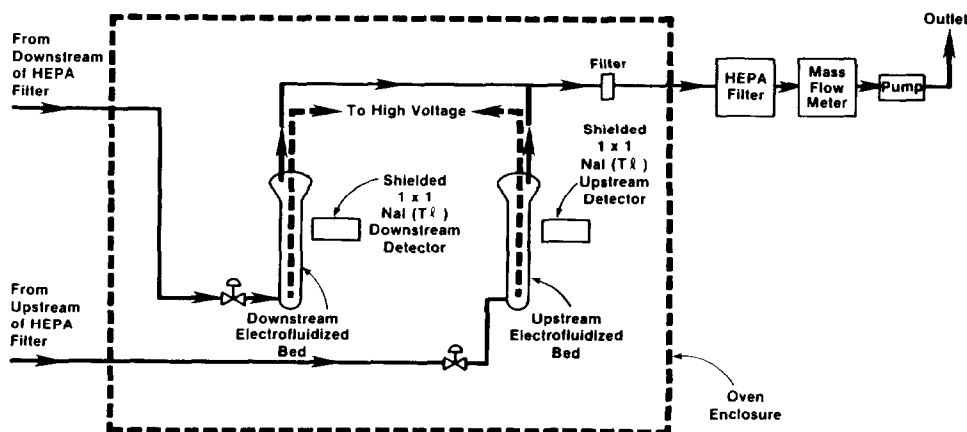


FIGURE 2 ON-LINE HEPA FILTER TEST APPARATUS

Laboratory Evaluation

The objective of the laboratory evaluation was two-fold: 1) To determine the ability of NaI(Tl) detectors (and associated electronics) to operate for extended periods of time at temperatures as high as 70°C; and 2) To determine the feasibility of operating the entire monitor (as shown in Figure 2) on a continuous basis. The achievement of these objectives is discussed below.

High Temperature Detector Operation

The maximum anticipated dew point at the Idaho Chemical Processing Plant (ICPP) Vessel Off-Gas (VOG) system is 60°C. Therefore, 70°C was selected as the design temperature for the proposed monitor to minimize the chance of condensation within the monitor. The NaI(Tl) crystals, photomultiplier tubes, and voltage distribution bases were placed within an oven used to maintain the detectors at the preselected temper-

ature. Because preamplifiers cannot be operated above 40°C, a two meter cable carried the signal to the preamplifier located outside the oven. The preamplifier and other supporting hardware were at ambient temperature.

The spectrum was accumulated with an ND60 multichannel analyzer (MCA). To determine the effect of high temperatures on the NaI(Tl) counting efficiency, a 9.13 μCi ^{137}Cs point source was counted at three temperatures (ambient, $40 \pm 2^\circ\text{C}$, and $70 \pm 2^\circ\text{C}$) and at five distances from the detector ($77 \pm 2\text{mm}$, $86 \pm 2\text{mm}$, $117 \pm 2\text{mm}$, $158 \pm 2\text{mm}$, and $204 \pm 2\text{mm}$). The detectors were allowed to equilibrate at each temperature overnight. At least 22,500 counts were accumulated at each temperature and distance, resulting in a 95% confidence interval of $\pm 1.5\%$ owing to counting statistics alone.

The five count rates measured at each temperature were normalized to the expected count rate at 10 mm by fitting to an inverse square law; These data are plotted in Figure 3. The temperature coefficient for the upstream detector was 0.605%/K; for the downstream detector it was 0.0894%/K. Since oven temperatures can be controlled to $\pm 2\text{K}$, the maximum error in the collection efficiency measurement due to temperature fluctuations would be $\pm 0.0012\%$, or 2.5% of the 0.05% penetration required to cause a HEPA filter to fail. This stability was considered suitable for most in-plant operations.

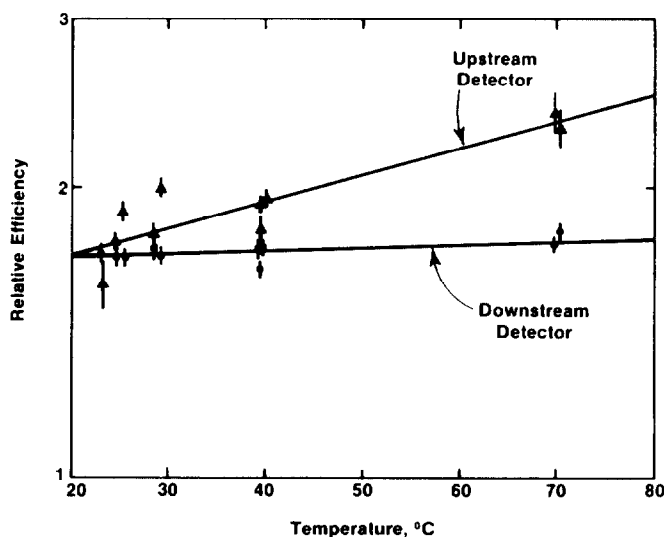


FIGURE 3 RELATIVE EFFICIENCY VS. TEMPERATURE

Integrated Monitor Operations

The second laboratory objective was to demonstrate long-term stability of the instrumentation and reliability of the integrated monitor during unattended operation. To accomplish this, the integrated HEPA filter monitor was assembled (as shown in Figure 4) and enclosed by a large oven to maintain the apparatus at a constant 70°C . A 47-mm Gelman Type A/E glass fiber filter simulated the HEPA filter to be tested. A portion of the challenge aerosol was continuously collected by the upstream EFB, the aerosol penetrating the test filter was continuously collected by the downstream EFB.

Two mass flowmeters measured the sample flows: the first was installed on the sample return line to measure the combined flowrate

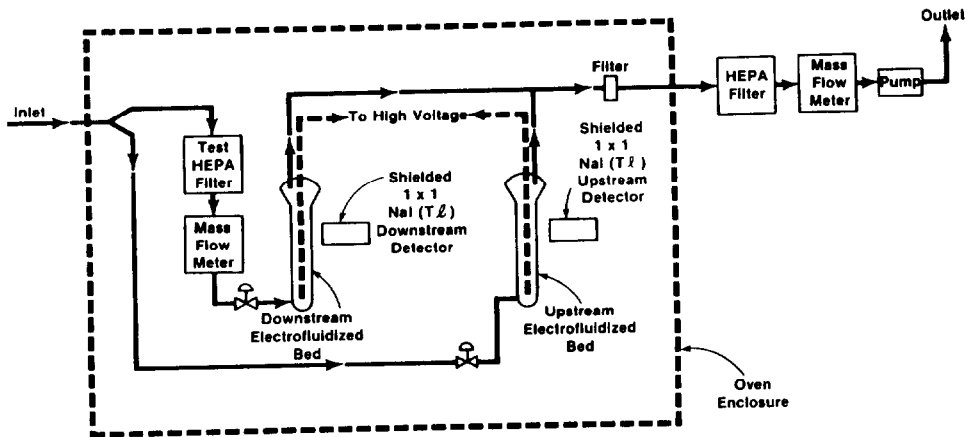


FIGURE 4 ON-LINE HEPA FILTER TEST APPRATUS

through both EFB's; the second, specially constructed for operation at temperatures up to 100°C, measured that portion of the total flow drawn through the test filter.

The NaI(Tl) detectors used to measure the particulate activity collected on the EFB's were shielded with 100 mm of lead. The detectors were further shielded with Mu metal to minimize the effects of magnetic fields generated by nearby motors. Output signals from the two detectors were processed simultaneously using an ND 564 gated analog router and an ND 60 MCA. Data accumulated by the MCA were periodically printed out.

Laboratory operation of the integrated monitor for approximately 24 hours over 3 days revealed the following:

- 1) Modification of the 220 V oven to operate at 110V permitted adequate heating power and temperature control for unattended operation;
- 2) A 6 mm x 12 mm Teflon sheath was adequate insulation for the 10 KV high voltage lead;
- 3) The sample flows were sufficiently stable to maintain adequate fluidization in the EFB's during unattended operation.

Therefore, since the laboratory operation had demonstrated that sustained operation of the NaI(Tl) at 70°C appeared feasible and that the assembled, integrated monitor could perform without difficulty, the monitor was installed at the ICPP Vessel Off-Gas (VOG) system for an in-plant demonstration.

In-Plant Demonstration

The in-plant demonstration of the integrated monitor was performed at the VOG sampling station. The off-gas at the VOG sampling contains radioactive particles from the tank farm, ILW evaporator, and steam jets. The typical particle size for this system is 0.05 μm before filtration and 0.015 μm after filtration¹. A block diagram of the pertinent features of the VOG system is shown in Figure 5.

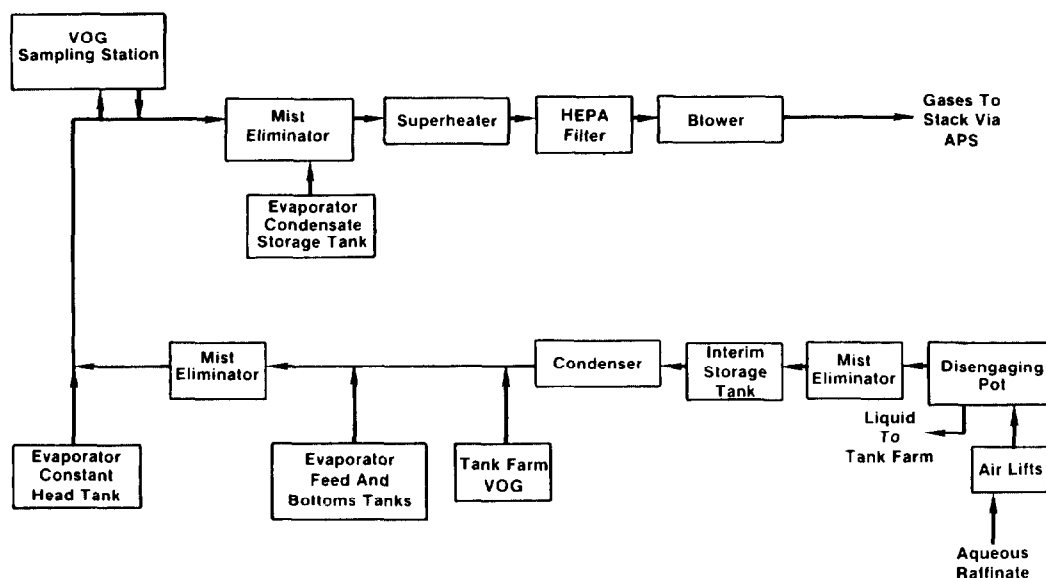


Figure 5. Block Diagram Of VOG System

Samples were withdrawn from the VOG line, passed through the prototype HEPA filter efficiency monitor, and returned to the VOG downstream. The 47 mm Gelman Type A/E glass fiber filter was again used to simulate the HEPA filter to be tested. The in-plant demonstration was performed in two parts: normal operation and abnormal operation.

Normal Operation

Normal operation is defined as the monitoring of a 99+% efficient filter in an off-gas stream with a dew point of less than 70°C. The integrated monitor was operated at the VOG system for 144 consecutive hours (with hourly printouts) of normal operation under the conditions shown in Table I. The results of this run are shown in Figure 6; Table II presents a statistical analysis of the data collected by the downstream detector. No significant penetration was observed when averaged over the 6 days, but significant slopes were observed on 4 of the six individual days. The source of this variability is unknown, but may be due to a combination of counting statistics, temperature variations, and fluctuations in the ambient gamma field.

Of greater interest is the HEPA filter efficiency measurement. The only times sufficient activity were challenging the test HEPA filter was during the two process transients indicated in Figure 6: Transient 1- During hours 23-24 a deep tank transfer to the evaporator feed tank of a 20 mR/cm³ solution was accomplished. During hours 30-32 a tank truck transferred a <0.001 mR/cm³ solution to the evaporator feed tank. Transient 2- During hours 64-67, a 500 mR/cm³ liquid waste was transferred to a deep tank.

The results of HEPA filter efficiency measurement during Transient 1 are shown in Table III. A detailed plot of the upstream response to Transient 1 is shown in Figure 7. As indicated in Table III, the sensi-

TABLE I

OPERATING CONDITIONS DURING NORMAL OPERATION

Parameter	Value
Ambient Temperature	10-23°C
Oven Temperature	70 ± 2°C
Total Sample Flow	4760 ± 50 cm ³ min ⁻¹
Challenge Sample Flow	1960 ± 40 cm ³ min ⁻¹
Applied Voltage	10 ⁶ V m ⁻¹
Ratio of Detector Efficiency (upstream/downstream)	1.93

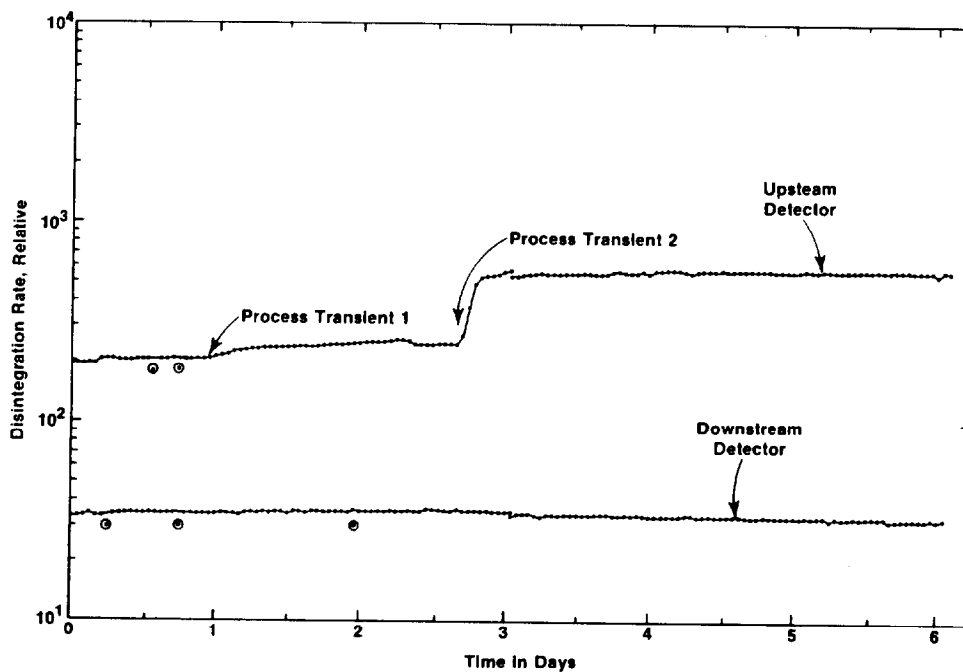


FIGURE 6 DISINTEGRATION RATE VS. TIME FOR HEPA RATED FILTER

ICPP-S-8507

TABLE II
STATISTICAL ANALYSIS OF DOWNSTREAM DETECTOR
DURING NORMAL OPERATION

Time After Experiment Start (h)	Number of Points	Average Count Rate (s^{-1})	Std Dev.	Slope (s^{-1} h^{-1})	95% Confidence Level of slope
0-23	22	34.39	0.441	+0.0500	± 0.0309
24-47	23	35.19	0.341	+0.0380	± 0.0223
48-71	24	35.75	0.198	+0.0098	± 0.0121
72-95	24	35.37	0.209	-0.0095	± 0.0128
96-119	24	34.48	0.243	-0.0197	± 0.0149
120-143	24	34.34	0.316	-0.0303	± 0.0193
0-143	141	34.99	0.576 ^a	-0.0028	± 0.0024

^a The observed average is $34.99 \pm 3.3\%$ whereas the standard deviation based on counting statistics is 0.8%.

TABLE III
HEPA FILTER EFFICIENCY MEASUREMENT DURING TRANSIENT 1

Time After Start of Experiment (h)	Detector	Number of Points	Slope ($s^{-1}h^{-1}$)	Correlation Coefficient	HEPA Filter Efficiency
15-43	Downstream	28	0.03527	0.7802	N/A
15-23	Upstream	8	0.1630	0.9252	>70.03%
24-30	Upstream	7	3.507	0.9989	>98.61%
31-43	Upstream	13	0.9012	0.9915	>94.56%

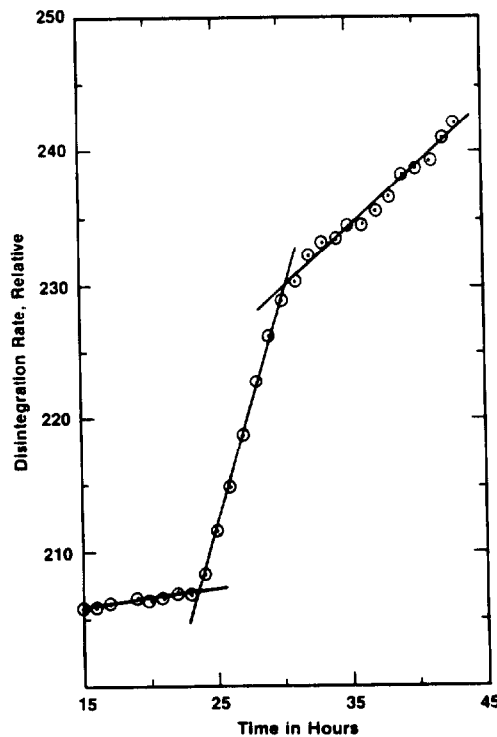


FIGURE 7 UPSTREAM DETECTOR RESPONSE TO PROCESS TRANSIENT 1

tivity of the monitor was limited by the small amount of activity released during this Transient.

The HEPA filter efficiency measurement was more sensitive during process Transient 2 because more than 10 times the activity was released during this period. A detailed plot of the upstream response to process Transient 2 is shown in Figure 8; the results of the HEPA filter efficiency measurement during process transient 2 are shown in Table IV.

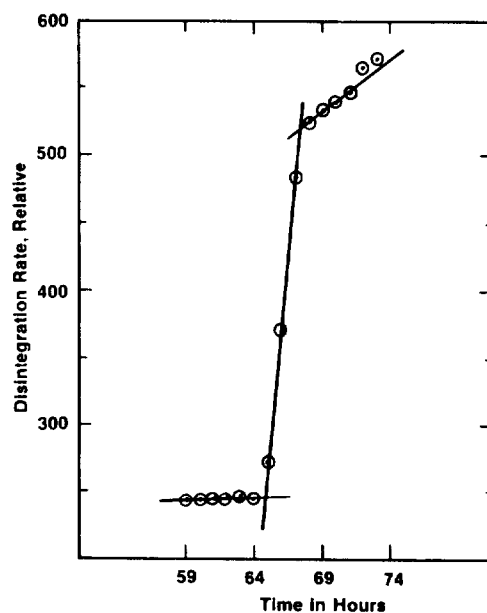


FIGURE 8 UPSTREAM DETECTOR RESPONSE TO PROCESS TRANSIENT 2

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From the data in Table IV it can be concluded that the integrated monitor is capable of monitoring a greater than 99% efficient HEPA filter under normal operation. Further, the fact that each transient is a common activity at a fuel reprocessing plant demonstrates that the integrated HEPA filter efficiency monitor has sufficient sensitivity to verify the integrity of HEPA filters during common activities.

TABLE IV
HEPA FILTER EFFICIENCY MEASUREMENT DURING TRANSIENT 2

Time After Start of Experiment (h)	Detector	Number of Points	Slope ($s^{-1}h^{-1}$)	Correlation Coefficient	HEPA Filter Efficiency
59-77	Downstream	18	-0.04223	-0.8624	N/A
59-64	Upstream	6	0.3457	0.8586	>99.05%
65-67	Upstream	3	105.35	0.9992	>99.99%
68-73	Upstream	6	3.400	0.5892	>99.90%

Abnormal Operation

Abnormal operation is defined as the monitoring of a breached filter or under conditions of excessive humidity or high concentrations of gaseous radionuclides. The operating parameters during abnormal operation are shown in Table V.

The objective of the breached filter experiment was to demonstrate that the monitor could rapidly detect a filter failure. Therefore, a 47 mm Gelman Type A/E filter was punctured to simulate a filter breach; the monitor was subsequently operated for 96 consecutive hours.

The data collected during this run are plotted in Figure 9. About 43 hours into the test, the weakened filter apparently permitted the release of some collected aerosol activity or contaminated filter material into the downstream EFB.

TABLE V
OPERATING CONDITIONS DURING ABNORMAL OPERATION

Parameter	Value
Ambient Temperature	10-23°C
Oven Temperature	70 ± 2°C
Total Sample Flow	4400 ± 110 cm ³ min ⁻¹
Challenge Sample Flow	2125 ± 135 cm ³ min ⁻¹
Applied Voltage	10 ⁶ V m ⁻¹
Ratio of Detector Efficiency (upstream/downstream)	3.02

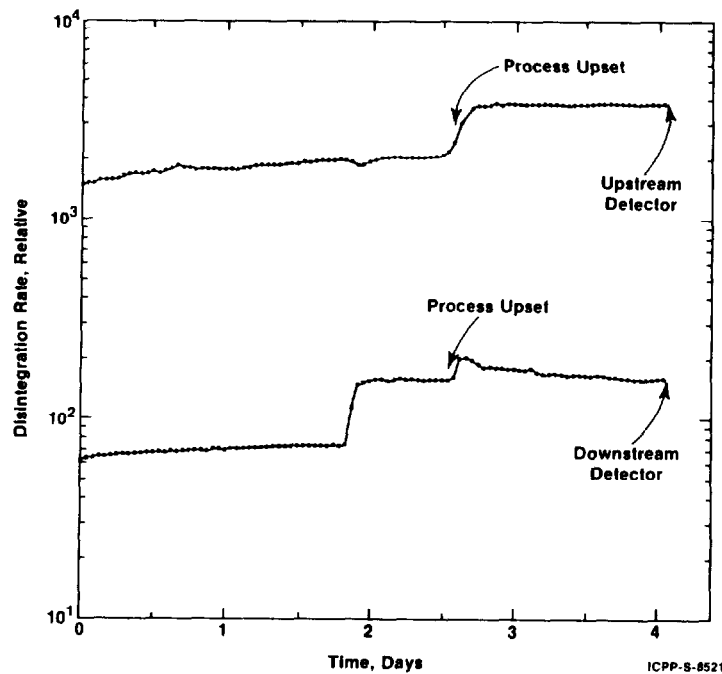


FIGURE 9 DISINTEGRATION RATE VS. TIME
FOR SIMULATED BREACHED FILTER

The process upset 61 hours into the test (also shown in Figure 9) was the consequences of two transfers of high-level liquid waste to the deep tanks. More detailed plots of the upstream and downstream detector responses during this process upset are shown in Figures 10 and 11.

The results of the filter efficiency measurements during this process upset are shown in Table VI. These data illustrate the ability of the HEPA filter efficiency monitor to detect breached at times of relatively high challenge aerosol radioactivity. The data also illustrate two potential drawbacks of EFB monitoring technology.

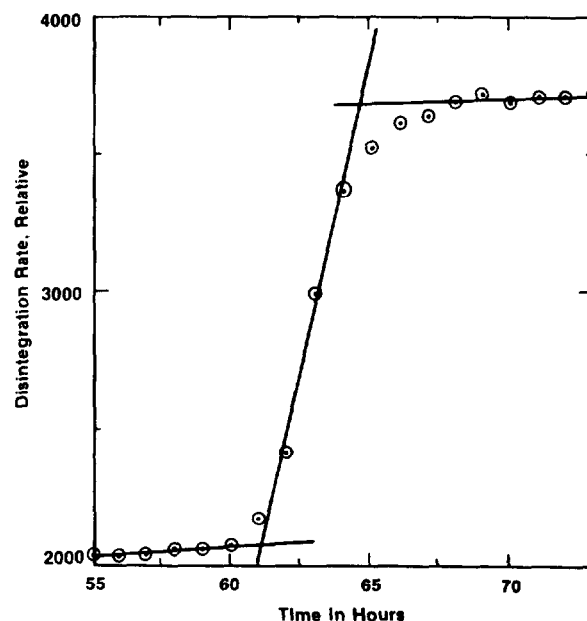


FIGURE 10 UPSTREAM DETECTOR RESPONSE
TO PROCESS UPSET

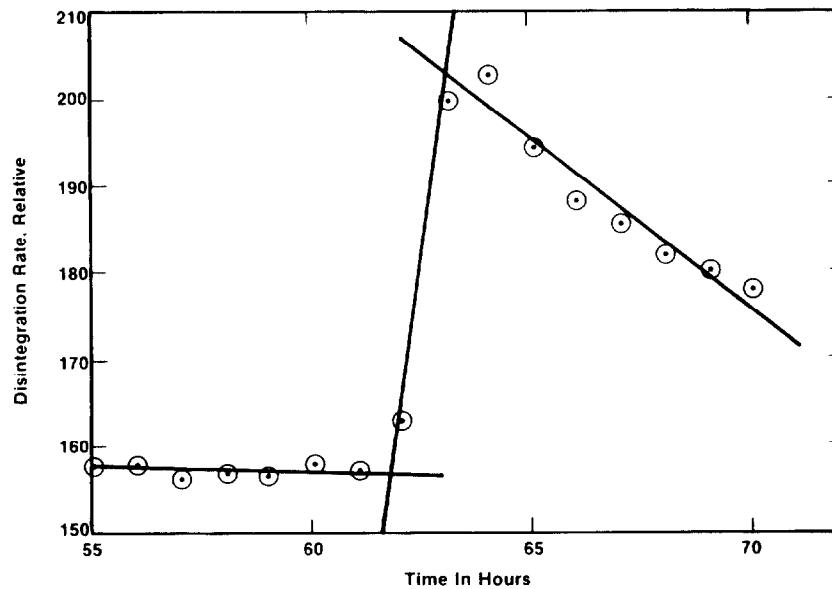


Figure 11. Downstream Detector Response To Process Upset

TABLE VI
HEPA FILTER EFFICIENCY MEASUREMENT DURING PROCESS UPSET

Time After Start of Experiment (h)	Detector	Slope (s ⁻¹ h ⁻¹)	Correlation Coefficient (r)	HEPA Filter Efficiency %
55-61	Downstream	-0.1107	-0.3340	N/A
62-63	Downstream	+36.80	1.000	N/A
64-70	Downstream	-38.57	-0.9612	N/A
55-60	Upstream	+9.371	0.9791	92.95%
61-66	Upstream	+310.7	0.9683	87.38%
67-72	Upstream	+10.49	0.71-9	N/A

The first drawback is the monitor's reaction to changes in fluidization during rapid pressure changes in the sample stream. The steep negative slope of the downstream detector during hours 65-68 (Figure 11) is attributable to poor fluidization (and thus poor mixing); the poor fluidization resulted from a rapid pressure transient within the fluidized bed. However, this drawback may be overcome by maintaining a constant pressure within the EFB.

The second drawback is the occurrence of a very slight (but significant at the 95% confidence level) negative slope of the downstream detector in the absence of penetrating aerosol activity. Although the cause of this effect is unknown, one explanation is the loss of fines from the bed which are transported to the back-up filter. Two other factors of the monitor's performance were evaluated during in-plant testing: the effect of condensation in the off-gas line, and the monitor's sensitivity to volatile forms of ¹⁰⁶Ru.

Effects of Condensation

Subsequent experiments encountered upset conditions at the VOG system resulting in condensation within the VOG duct. This condensation then flooded the electrofludized beds. Routine operation was resumed after operating the monitor on ambient air for 24 hours to dry the system. No external contamination was encountered during this operation.

 ^{106}Ru Sensitivity

The effect of gaseous $^{106}\text{RuO}_4$ on the HEPA filter efficiency measurement was also evaluated. If the NaI(Tl) is unable to distinguish between ^{106}Ru and ^{137}Cs , the presence of $^{106}\text{RuO}_4$ would cause an intact HEPA filter to be classified as breached. Therefore, a 2^3 factorial design experiment was conducted in duplicate to determine the relative counting efficiency of the NaI(Tl) for ^{106}Ru when the region of interest is centered on the ^{137}Cs 662 KeV gamma peak.

Three sealed sources were used in this experiment: a 2.49 μCi ^{106}Ru source, a 1.76 ^{137}Cs source, and a 9.13 μCi ^{137}Cs source. Each source or combination of sources was attached to the EFB opposite the NaI(Tl) and the count rate recorded. The 2^3 factorial design allowed compensation for background and backscatter effects. The results of this experiment are shown in Table VII.

As is shown by the data in Table VII, there was no significant effect due to the presence of ^{106}Ru . If one calculates the maximum effect based on the uncertainty of the measurement, then the ratio of the counting efficiency for ^{106}Ru to the counting efficiency of ^{137}Cs would be <0.24 . Therefore, as long as the gaseous $^{106}\text{Ru}/^{137}\text{Cs}$ ratio is less than $(.0005/0.24)$ or 0.002, the presence of ^{106}Ru will not cause an intact filter to measure less than 99.95% efficiency. Because this condition is met at most nuclear facility off-gas streams, no further measures were taken to lessen the response of the monitor to ^{106}Ru .

TABLE VII
RESULTS OF ^{106}Ru INTERFERENCE EXPERIMENT

Source Present	Counting Effect (cps/ μCi)
2.49 μCi ^{106}Ru	4.94 \pm 7.07
1.76 μCi ^{137}Cs	61.14 \pm 10.00
9.13 μCi ^{137}Cs	52.62 \pm 1.93
10.89 μCi ^{137}Cs	56.19 \pm 1.62
2.49 μCi ^{106}Ru + 1.76 ^{137}Cs	77.16 \pm 10.00 ^a
2.49 μCi ^{106}Ru + 9.13 ^{137}Cs	52.19 \pm 1.93 ^a
2.49 μCi ^{106}Ru + 10.89 μCi ^{137}Cs	55.15 \pm 1.63 ^a

^a Based on ^{137}Cs present

Conclusions

A technique was developed and demonstrated for the continuous monitoring of HEPA filter efficiencies by simultaneously measuring the challenging and penetrating radioactive aerosol concentrations. The upstream and downstream measurements are accomplished by quantitative trapping of all 0.002 - 0.200 μm particles on electrofluidized beds. The collected aerosol is continuously measured by gamma spectrometry.

The technique is an improvement over the three commonly used methods: it is an improvement over the French uranine method because prompt, on-line results are available. Unlike the United Kingdom sodium chloride method, it is useful at higher humidities and does not introduce potentially corrosive salts. And, further it is better than the American DOP method because no assumptions regarding the particle size distribution are necessary - the measurement is made on the actual aerosol of interest.

The major conclusions of this work are:

1. The monitor is sufficiently sensitive to measure the efficiency of a 99.95% efficient filter under normal conditions provided there is some plant process operating;
2. Process upsets, process transients, and filter breaches are accurately detected;
3. Acceptable reliability was demonstrated under in-plant conditions and rapid recovery from condensation in the off-gas system are afforded;
4. Selectivity is limited only by the possible presence of very high concentrations of gaseous ^{106}Ru in the off-gas.

Accordingly, the in-place, realtime HEPA filter efficiency monitor is suitable for most nuclear facility off-gas applications.

References

1. F. A. Hohorst and S. J. Fernandez, "Development of Submicron Particle Size Classification and Collection Techniques for Nuclear Facility Off-Gas Streams," ENICO-1076 (1981).
2. S. J. Fernandez, G. D. Pierce, L. P. Murphy, and B. G. Motes, "Evaluation of Submicron Particle Size Classification and Collection Techniques for Nuclear Facility Off-Gas Streams," ENICO-1003 (1979).

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DISCUSSION

COOPER: What are the advantages of electrofluidized beds as samplers in comparison to electrostatic precipitators, thermal precipitators, or even absolute filters?

HOHORST: An electrofluidized bed was deemed more suitable for remote operation. In addition, a uniform geometry could be obtained to assist in achieving precise radioactivity measurements with NAI detectors.

ETTINGER: What uncertainty would you assign to the calculated 99.9% HEPA filter efficiency based on the uncertainty in the measured slope and air flow rate?

HOHORST: I don't believe I can give you a fixed number as the answer would relate to what the downstream filter measurement is. That would govern the calculation.

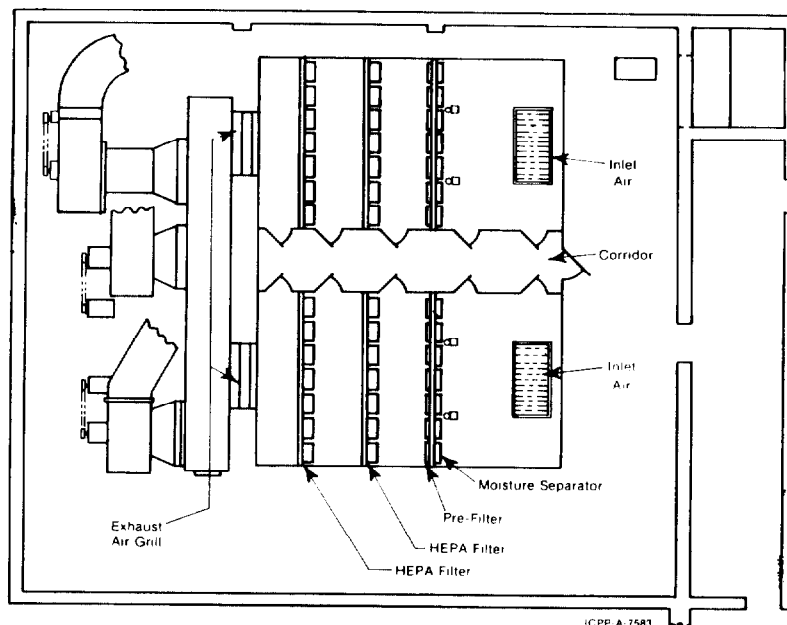
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DOP TESTING HEPA FILTER BANKS IN SERIES

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Abstract

A new method has been developed to DOP test large HEPA filter banks installed in series. In two (2) different systems containing 28 HEPA filters and 42 HEPA filters respectively, each having another bank of HEPA filters in series, a system was installed to provide a uniform concentration of DOP smoke within a 5' space that exists between banks. (Figure 1)



Calciner Exhaust Air Plenum Room

Figure 1

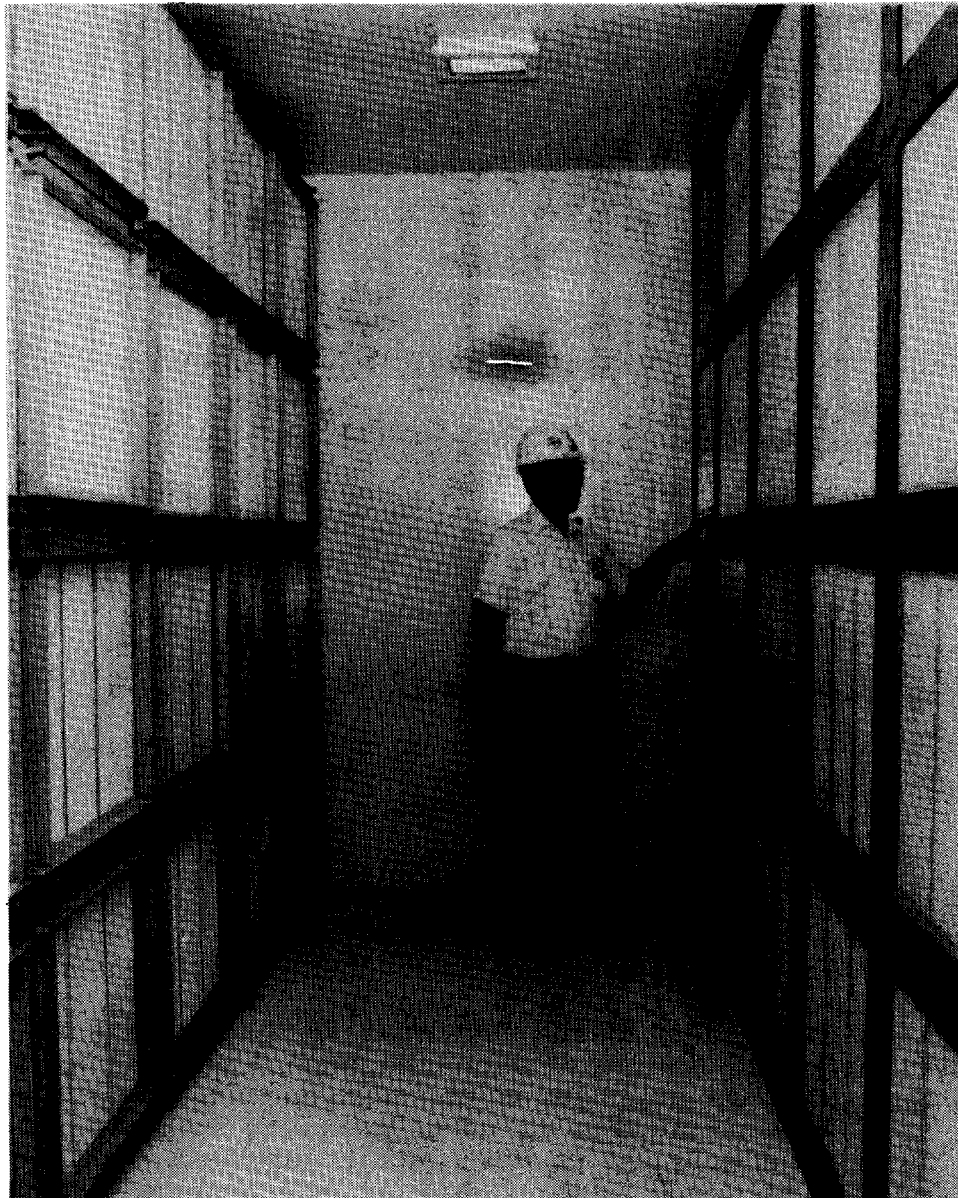
I. Introduction

The New Waste Calcining Facility (NWCF) is located at the Chemical Processing Plant (CPP) at the Idaho National Engineering Laboratory (INEL). It is being operated by EXXON Nuclear Idaho Company (ENICO) under contract with the Department of Energy (DOE). This facility calcines and stores the radioactive liquid waste resulting from separating the fission products from the enriched uranium. This new facility is designed to approximately double the capacity of its predecessor. All of the exhaust systems are filtered by a 5½" thick ACS moisture separator, a 35% pre-filter, and two stages of HEPA filters before being discharged to the atmosphere. In addition, all of the supply air is HEPA filtered along with additional HEPA filters before it is introduced into each

cell. There are approximately 425 HEPA filters in the NWCF H & V systems.

II. Initial Design and Sealing Problems

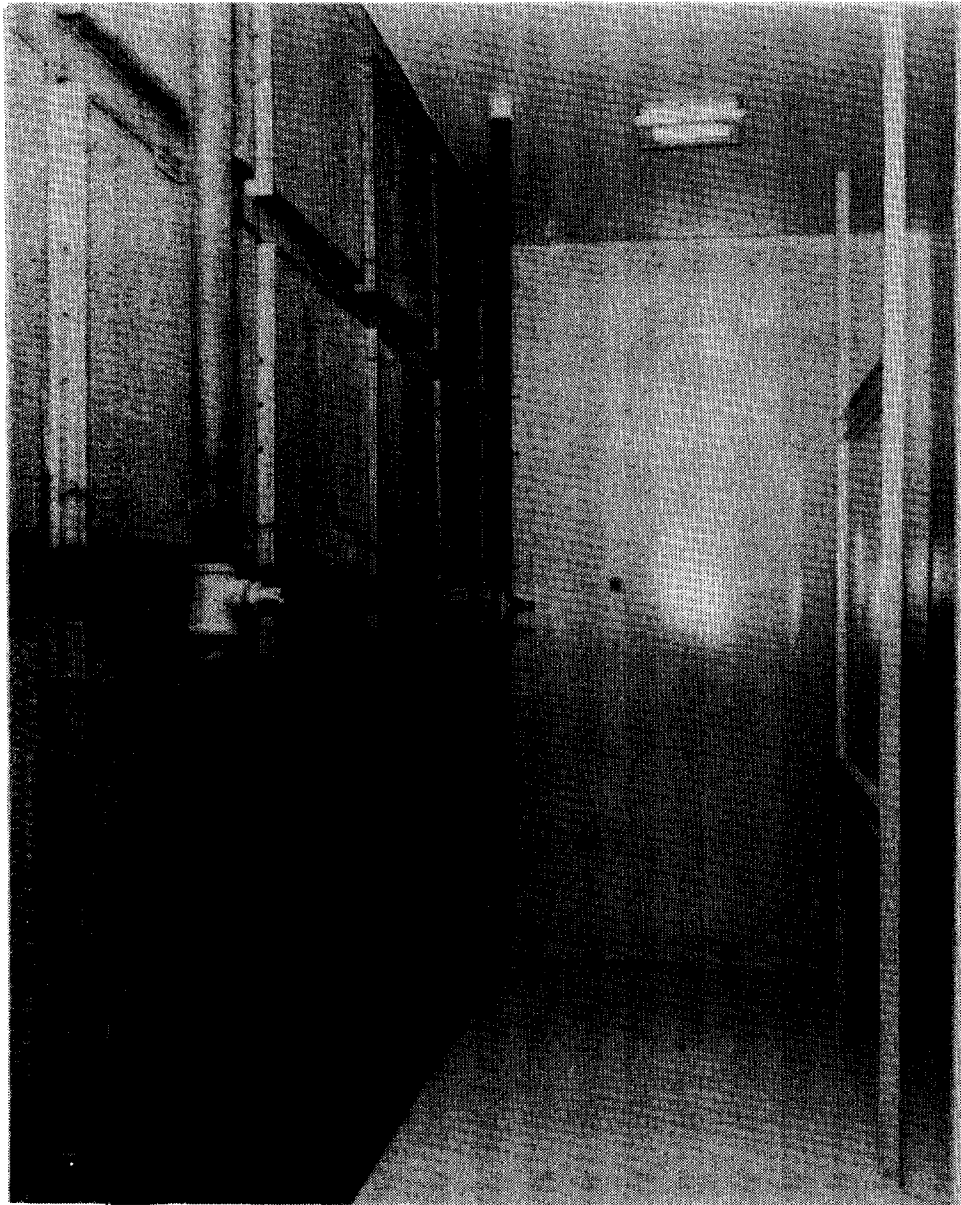
There are two main exhaust systems in the facility other than the calciner off-gas cell. One provides HEPA filtration for the decontamination area and the other provides HEPA filtration for the balance of the facility. Originally the filters were of the gasket seal type. The sealing frame in the filter plenum was a stainless steel plate which had openings cut out approximately $22\frac{1}{2}$ " X $22\frac{1}{2}$ " square for the filters. (Figure 2)



**Downstream Of 1st Bank Of HEPA's And Upstream Side Of
2nd Bank**

Figure 2

In the general building exhaust systems, there are 42 HEPA filters in a single bank. There are two banks in series. The HEPA filters are preceded by a 35% pre-filter and a 5½" thick ACS moisture separator. (Figure 3) The moisture separators are intended to protect the HEPA filters in case the fire protection water spray system is actuated. There is a standby system which also has 42 filters per bank and is intended to be used during filter change-out and as a back-up should the other system fail. These two exhaust systems account for 280 of the 425 HEPA filters used in this facility.



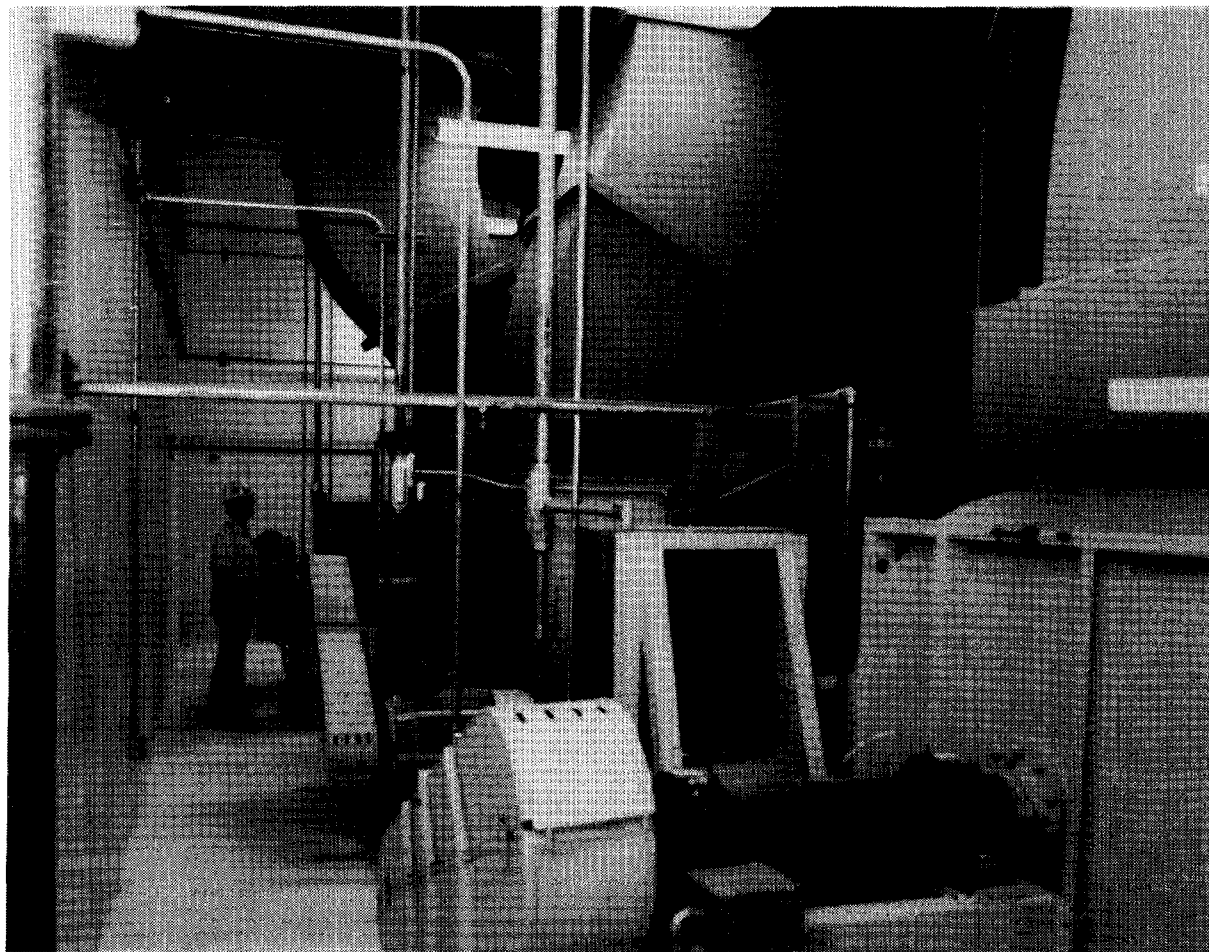
Moisture Separators

Figure 3

Initially there was no means provided to DOP test the 2nd bank of HEPA filters without removing the 1st bank. This practice is not desirable after the system becomes contaminated. After removing the 1st bank of filters the 2nd bank was sampled for the downstream concentration as it exited the plenum. The filters for the 1st bank were then re-installed and tested.

The 1st and 2nd banks were scanned approximately 2 inches from the filter. When scanning the 2nd bank (with the 1st bank of HEPA filter removed) numerous penetrations (up to 12% DOP) were detected.

However, at a point just downstream from the blower, it did not show DOP penetration above 0.03% which satisfies the ANSI N-510 1980 test requirement. (Figure 4) In light of today's stricter requirements, it seemed inconsistent to accept the results of a test performed in this manner. In this case there are two HEPA filters in series but the same amount of leakage is acceptable for a single bank. It was felt that every effort should be made to eliminate the major leaks as far as is practicable.



Calcliner Exhaust Fan Arrangement

Figure 4

III. Modifications Implemented

Obtaining an acceptable sealing surface was found to be impossible due to irregularities in the sealing frame surface so it was decided to convert the gasket type filters to a fluid seal type. (Figure 5) Knife edge adapter frames were purchased and installed. Subsequent scanning by the method used earlier showed no leakage greater than 0.20% DOP.



HEPA FILTER SYSTEM AFTER CONVERSION
TO FLUID SEAL WITH DOP DISTRIBUTION SYSTEM INSTALLED

Figure 5

Since this method of scanning is time consuming and ineffective, two devices were constructed which take into consideration pinhole leakage. One is a type of hood which is placed over the individual filter with a sample line going to the photometer. It is much faster and more representative of the filter leakage. (Fig.6)



Figure 6

The other device is a tapered hood 4" wide and 14" long. Other than a sample line which goes to the photometer, it is enclosed. (Figure 7) It has an open face that is held 6" away from the filter by means of an arm with a roller. The unit is operated by simply rolling down the face plate of each filter section. It is somewhat easier to spot small leaks than the first hood. Any penetration less than 0.1% is considered acceptable.

A bank of 42 filters can be scanned by employing one of the two hood methods in approximately 20 minutes compared to approximately 3 hours using the original scanning method.

Since there was no approved method for introducing and mixing the DOP smoke for the 2nd HEPA filter bank, a system to be used for each HEPA filter bank in each of the plenums was designed. This system essentially consists of a main header (3" pipe) installed horizontally in the middle of the downstream side of the preceding filter bank. Vertical 3/4" pipes are connected to the header and spaced between every 2nd filter. Each 3/4" pipe has 3/32" I.D. orifices positioned so that the flow through the orifice is perpendicular to the air flow through the filter. There is an orifice every 4" on both sides of each of the 3/4" branch lines to provide for uniform distribution vertically from the top to the bottom of the plenum. There is a 5' space between each filtration stage.

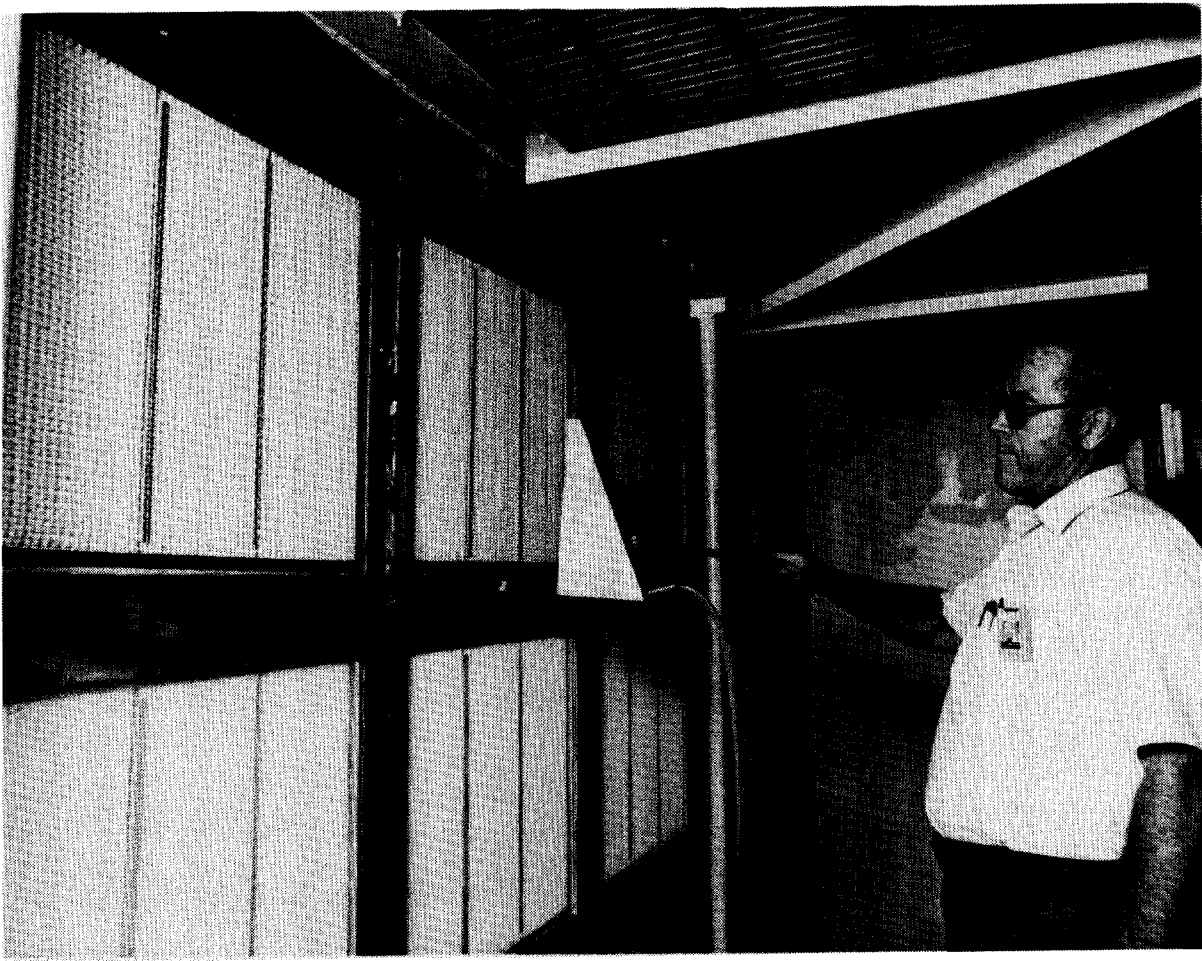


Figure 7

The main 3" header penetrates the wall of the filter housing which is connected to a Penberthy-Howdaille air jet pump. (Figure 8) A 1" I.D. plant air line is connected to the fitting on the jet pump and a flexible 2" I.D. hose is attached to the suction side of the jet pump with the other end loosely fitting over the discharge opening of the thermal DOP generator. When the air pressure is turned on, it creates a suction which draws in the DOP smoke. The air pressure forces the smoke through the orifices in the 3/4" piping and distributes the smoke over the width of each filter, providing a uniform smoke pattern to challenge each filter stage. Tests show that DOP concentrations were maintained within $\pm 5\%$ anywhere along the filter plane. It has also proven to be important to calculate the total area of all the orifices so that they are nearly equal to the area of the header. If they are too small, suction will not be created by the jet pump. If they are too large, the flow may not be uniform within the plenum. Care must also be exercised to make certain the orifices are perpendicular to plenum flow in order to obtain best results.

Here at the ICPP, the jet pump principle has been used for several years in systems where it is impossible to achieve adequate mixing. In this case the jet pump is connected to a 3/4" pipe having many more orifices and inserted into a duct.

IV. Other Areas of Interest

The material used for DOP distribution piping, inside the plenums was PVC, since the atmosphere is slightly corrosive. Two or three weeks after the piping was installed and DOP tests were completed it was observed that most of the piping was shattered and was laying on the floor of the plenums. Investigation has revealed that DOP will cause PVC embrittlement, which occurred even though there had been no pressure in the system except during the DOP test. The PVC piping is being replaced with stainless steel at the present time.

Careful consideration should be given for the proper design of filtration systems. The basic filter plenum requires two sets of HEPA filters, a pre-filter and a moisture separator. The entire system has to be duplicated to allow changeout of filters and provide a back-up system, whereas if bag-out housing had been used the only duplication required would have been a filter housing and possibly a fan. The advantages of bag-out housings are; more precise testing, reduced space needed for equipment, better contamination control, no need to enter the housing during the filter testing and change-out of filters, reduced cost of equipment and installation, fewer number of people needed for testing and filter change etc.

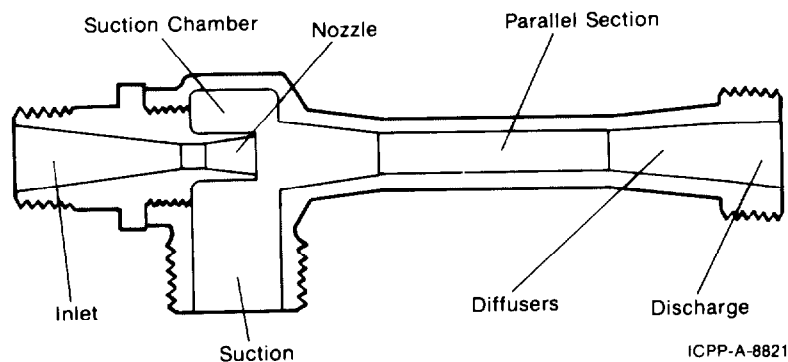
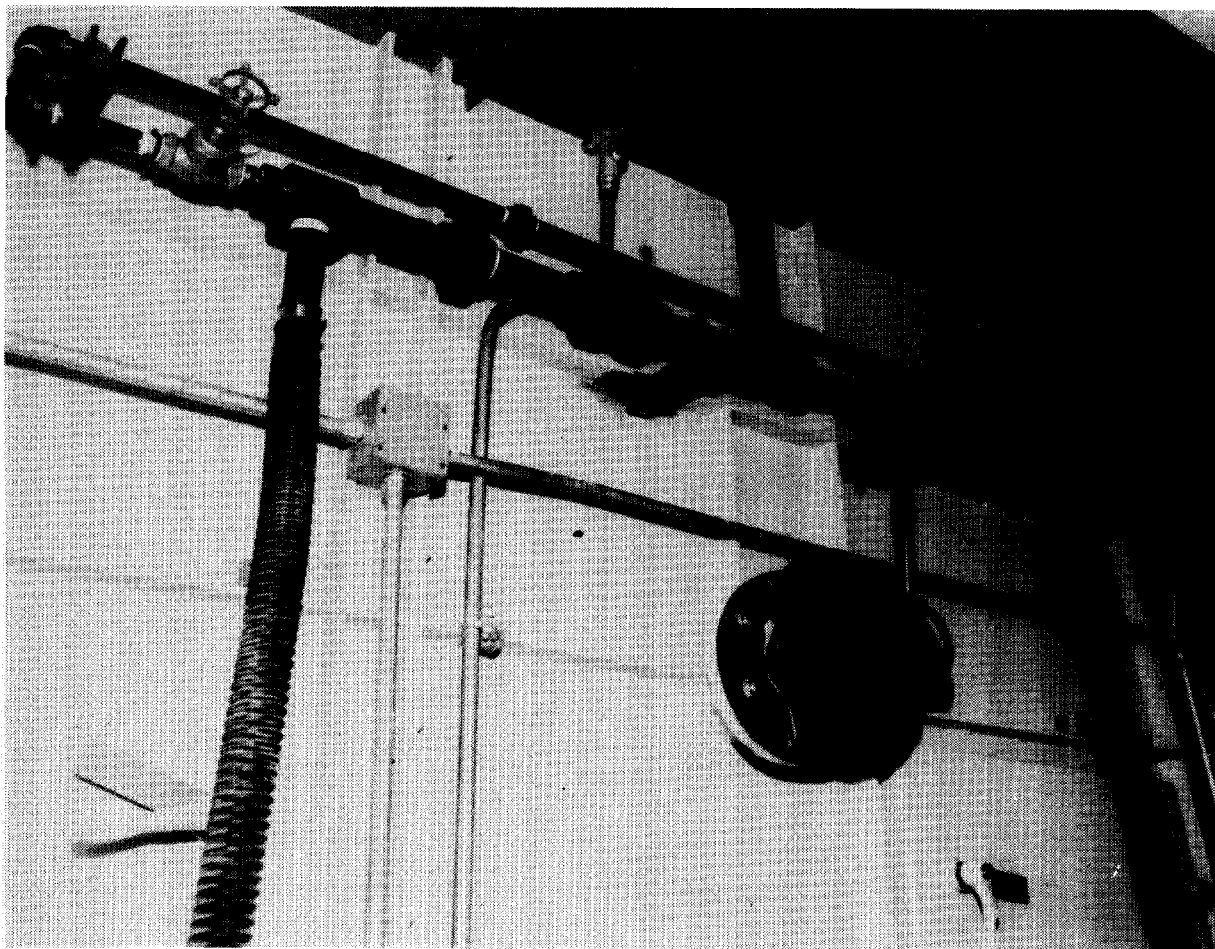


Figure 8



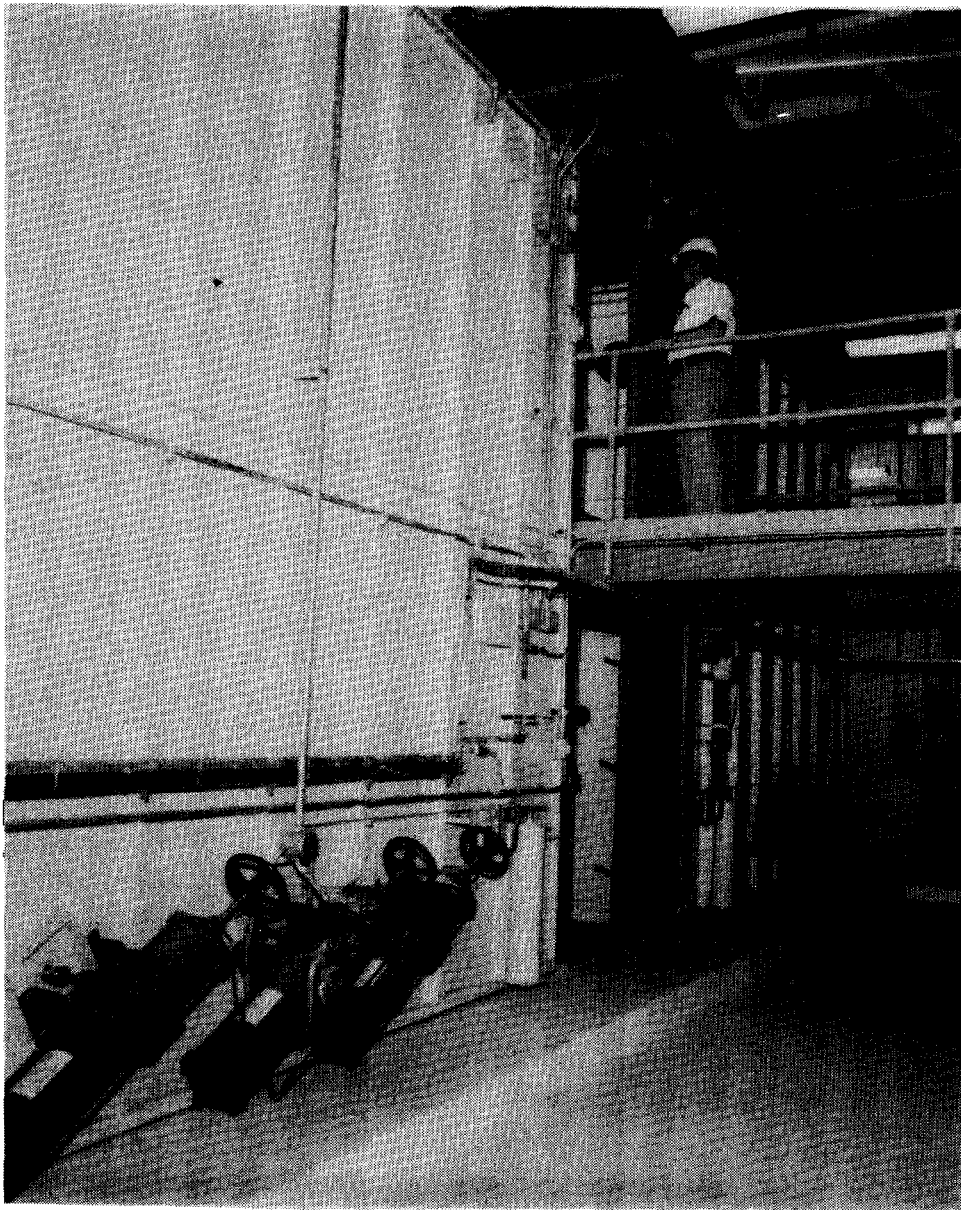
DOP INJECTION SYSTEM WITH JET PUMP

Figure 9



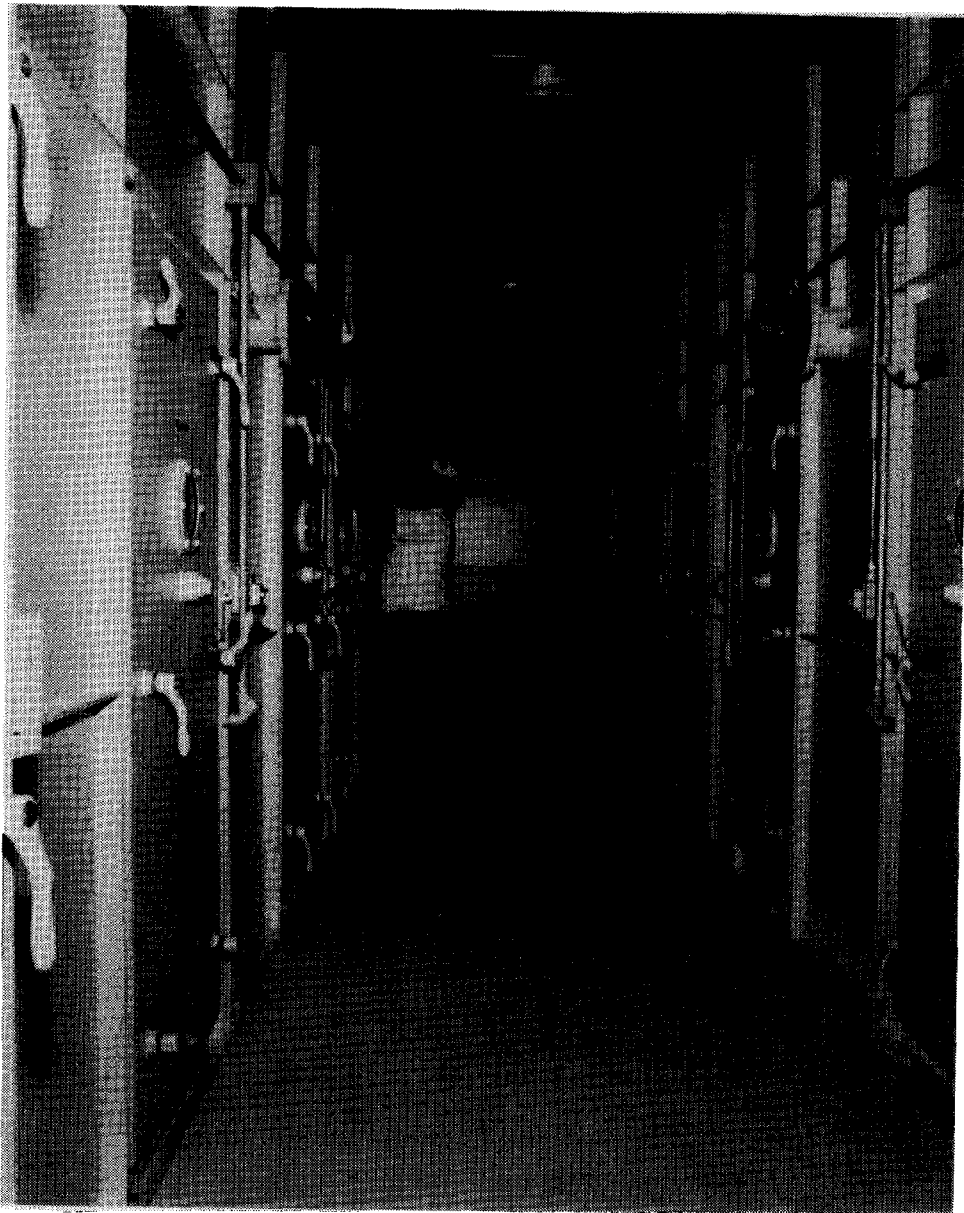
Exhaust Grill And Downstream Side Of 2nd HEPA Filter Bank

Figure 10



Calclner Exhaust Plenum Access To Both Levels

Figure 11



Calciner Exhaust Corridor Between Plenums

Figure 12

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DISCUSSION

JOHNSON, J.S.: What were the scanning results after the bank was converted to a fluid seal? Were leaks found along the two sides of the filters? Were there any consistent leak areas found when the banks were scanned?

HANSON: Leaks were eliminated and only low levels of pinholes, i.e., 0.1 to 0.2%, were observed.

ANON: What type of DOP generator was used and how many are required for a test?

HANSON: The ATI thermal generator was used. Two generators were used for the 42 filter bank but one generator would be sufficient if it is operating correctly.

WATSON: You said that a bank of 42 filters would be scanned by the manual roller/sampler in 20 minutes. Is that for active operation with a suited operator?

HANSON: The scanning itself took 20 minutes. Suiting up would add time as well as other anti-contamination controls.

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THE EFFECT OF PARTICLE SIZE VARIATION ON FILTRATION EFFICIENCY MEASURED BY THE HEPA FILTER QUALITY ASSURANCE TEST*

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Abstract

Satisfactory performance of new high efficiency particulate air (HEPA) filters is verified by quality assurance (QA) tests conducted prior to filter installation. Filtration efficiency for this test is defined as the ratio of the downstream aerosol concentration to the concentration upstream of the filter, as measured with a light scattering photometer (penetrometer) sensing light intensity scattered in the near forward direction. Existing QA acceptance criteria is interpreted as requiring the HEPA filter to be at least 99.97 per cent efficient for 0.3 μm diameter, unit density particles. The actual meaning of QA test results is dependent on a number of variables including the average size and size distribution of the challenge aerosol, the efficiency of the filter with respect to particle size, the size distribution of the penetrating aerosol, and the sensitivity of the photometer with respect to particle size and concentration.

A theoretical model has been used to calculate the filtration efficiency that would be indicated by the photometer for challenge aerosols of different size distributions and HEPA filters with different efficiencies as functions of particle size. The model compares the calculated overall efficiency indicated by the photometer with efficiencies calculated with respect to particle number and mass. This calculation assumes three aerosol distributions previously measured at the Filter Test Facilities (FTF) and four different filtration efficiency versus size curves.

The differences in efficiency measured by the QA test procedure and the efficiencies with respect to aerosol mass and number have been calculated for a range of different size particles. The results of these calculations are discussed.

I. Introduction

The measurement of air filter efficiency requires a challenge aerosol and instrumentation to determine aerosol concentration upstream and downstream of the filter. Filtration efficiency may be determined with respect to aerosol mass, particle number or any parameter for which the concentration measurements can be made. In general, filtration efficiency is expressed in terms of the ratio of the downstream concentration to the upstream concentration using the following relationship:

*Work performed under the auspices of the United States Department of Energy, Nuclear Fuel Cycle and Waste Management Division.

$$E = 100 \left(1 - \frac{C_D}{C_U} \right), \text{ where} \quad (1)$$

E is the efficiency in per cent, C_D the downstream concentration and C_U the upstream concentration. This calculation assumes that equivalent concentration measurements have been made at each location.

Quality Assurance (QA) or acceptance tests are conducted on High Efficiency Particulate Air (HEPA) filters to verify that the filters satisfy minimum performance criteria.⁽¹⁾ These tests should be carried out rapidly; should not impact on the useful life of the filter by heavy loading; and should use an aerosol material that does not alter the filter. The aerosol concentration measuring instrument must provide rapid or real time measurements and have sufficient sensitivity to verify acceptable performance. For most applications it is only necessary to verify that the filter efficiency is greater than some minimum value. In the case of HEPA filters, this is generally accepted to be an efficiency of 99.97 per cent when the filter is challenged by a 0.3 μm diameter test aerosol. The requirement for a rapid measurement for both high (upstream) and low (downstream) aerosol concentrations limits the types of satisfactory instruments. These requirements also make it more difficult to insure that equivalent concentration measurements are obtained at both sampling locations.

There are two common causes of nonequivalent concentration measurements. The large difference in concentration ($\sim 10^4$) encountered across HEPA filters is beyond the linear range of many instruments. In principle, this problem can be handled by careful instrument calibration. However, there is always some doubt about the general application of any calibration. A second cause of nonequivalent concentration measurements is the use of instruments whose response is particle size dependent. Instruments of this type may be used to measure filtration efficiency if the particle size distribution is known at both locations and appropriate corrections are made, or if the challenge aerosol is monodisperse. The existing HEPA filter QA test⁽²⁾ specifies the use of a monodisperse aerosol to insure that the size distribution of the upstream and downstream aerosols are identical and that concentration measurements are equivalent.

The monodisperse aerosol generator used for the FTF QA test is of the LaMer type⁽³⁾ where monodisperse aerosols are produced by evaporation of an organic material followed by controlled condensation of the material. In order to grow to the same size, all particles must have the same temperature and vapor concentration history. These conditions are difficult to obtain at the flow rates and concentrations required to conduct QA tests. Field studies reported elsewhere at this meeting⁽⁴⁾ indicate that the aerosols produced at the QA facilities are not monodisperse, but have geometric standard deviations greater than 1.35. This study has been carried out to investigate the effects of using a polydisperse aerosol with concentration measuring instruments designed for use with monodisperse aerosols.

II. Objectives and Approach

An analytical study was performed to address the potential error introduced in HEPA filter efficiency measurements by the determination of aerosol concentration using a penetrometer, whose response is particle size dependent, under test conditions where the aerosol upstream and downstream from the HEPA filter is polydisperse and has a different size distribution. The analytical study involved:

- a) Calculation of the HEPA filter efficiency defined by the penetrometer for three different assumed aerosol size distributions and four different assumed models of HEPA filter efficiency as a function of size.
- b) Calculation of the HEPA filter efficiency in terms of aerosol mass and particle number for the same aerosols and filter efficiency models noted in (a).

The assumed aerosol size distributions were selected to approximate the aerosols recently measured at the FTF.⁽⁴⁾ HEPA filter efficiency as a function of particle size was based on either of two experimental results or was adjusted from these experimental data to the QA acceptance criteria (99.97 per cent efficient for 0.3 μm diameter particles).

III. Aerosol Concentration Measurement Using a Photometer

The concentration measuring instrument specified for HEPA filter QA tests is the forward light-scattering photometer. This instrument will measure concentration over wide ranges and provides rapid determination of satisfactory filter performance. Figure 1 is a schematic view of a forward light-scattering photometer of the type used in the QA tests. The light from an incandescent lamp is focused in the center of a chamber containing the aerosol sample. The center of the lens is masked to form a conical annulus of light around a central shadow. If there are no particles present in the sample, then the pattern will be repeated beyond the focal point resulting in a shadow on the light-gathering lens system of the light detector. The presence of particles in the focal plane of the lens will result in the scatter of light into the conical shadow beyond the focal plane. This scattered light will be detected by the photomultiplier tube. The signal from the photomultiplier tube is converted to a concentration measurement by calibration of the system.

Dependency of the forward light-scattering photometer on particle size and index of refraction is well known. Sinclair proposed the use of this type of instrument for measuring particle size.⁽⁵⁾ The relative responses of this photometer as a function of particle size for di-ethylhexyl phthalate (DEHP) and di-ethylhexyl sebacate (DEHS) were calculated based on the geometry of a commercially available light-scattering photometer and the wavelength spectrum of the illuminating light which was determined by assuming the lamp is a black body radiator at a temperature of 2850° Kelvin. A Mie computer code was then used to calculate the amount of light scattered to the detector over the wavelength spectrum of the illuminating light. The sensitivity of the photomultiplier tube as a function of wavelength

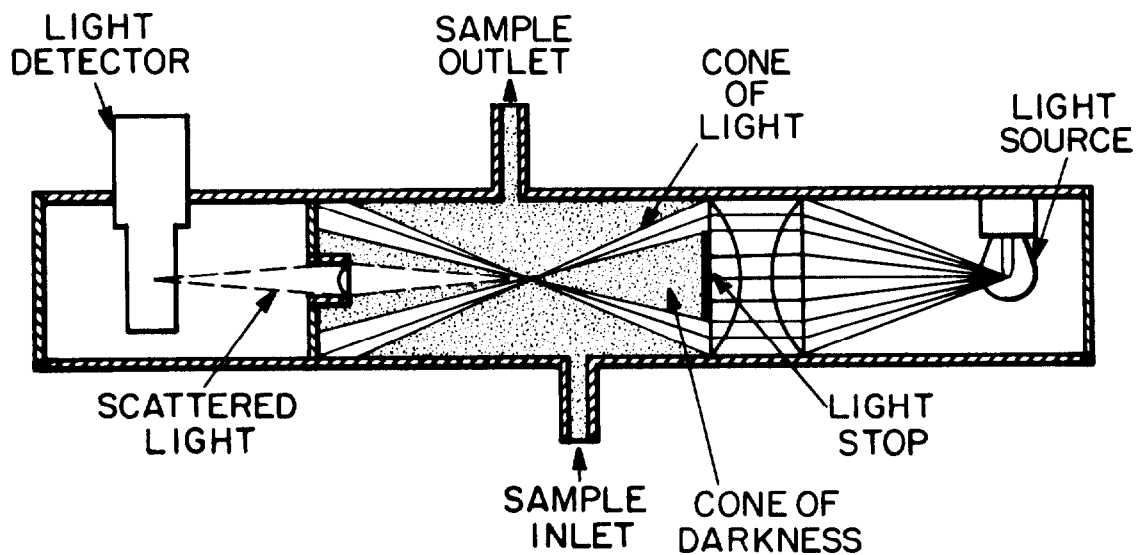


FIGURE 1
SCHEMATIC VIEW OF FORWARD LIGHT-SCATTERING PHOTOMETER

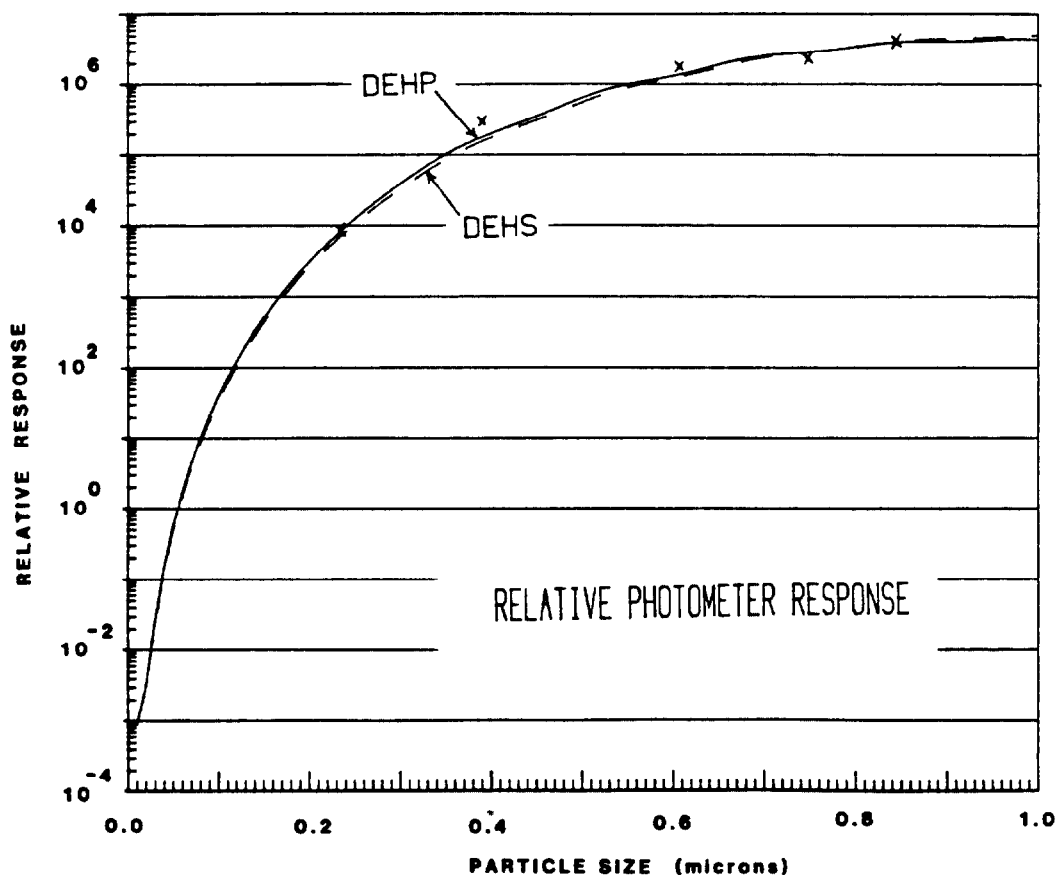


FIGURE 2
CALCULATED RELATIVE RESPONSE OF FORWARD LIGHT-SCATTERING PHOTOMETER
AS A FUNCTION OF PARTICLE SIZE FOR DI-ETHYLHEXYL PHTHALATE AND
DI-ETHYLHEXYL SEBACATE

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was used to determine the relative response of the photometer. Calculations were made for 100 particle diameter increments of 0.01 μm , for particles ranging in size from 0.01 μm up to 1.0 μm . The curve shown in Fig. 2 is the smooth connecting curve for these 100 calculated data points. Over the particle size range from 0.01 to 1.0 μm diameter, the relative response varies almost 10 orders of magnitude. The two curves for DEHP and DEHS are close together or coincide over this size range. The plotted points (x) on Fig. 2 are data presented by Sinclair⁽⁵⁾ as measurements of the voltages produced by the photomultiplier when particles of various sizes and refractive indices are passed through the photometer. These data were normalized to fit the curve at 0.84 μm and indicate good agreement between the calculated curve and the experimental data.⁽⁵⁾

Figure 2 illustrates the importance of particle size to the signal produced by the photometer. The curve indicates that it takes about one hundred 0.10 μm diameter particles to produce the same signal as one 0.20 μm diameter particle. The strong dependence of photometer response on particle size indicates that errors may result when concentrations, measured from aerosols having different size distributions, are compared on the basis of the signal obtained using this type of photometer. This effect can be important when measuring the efficiency of a HEPA filter using a polydisperse aerosol, where the size distribution of the test aerosol upstream and downstream from the filter are different.

IV. Procedure

The differences between filtration efficiency with respect to aerosol mass, aerosol particle number, and light-scattering photometer readings have been investigated by computer calculations. The calculations have been limited to the narrow range of filter test conditions encountered during QA tests of HEPA filters. A flow diagram indicating the steps used in this calculation is given in Fig. 3. The first step involved the numerical creation of an aerosol based on selected parameters for a log normal size distribution of particle volume. The size distribution was then sub-divided into 1700 intervals and the mass and number of particles in each interval was calculated using the log normal distribution function to determine mass and

$$N_i = \frac{6 M_i}{\rho \pi D_i^3} \quad (2)$$

to determine number. In equation (2): N_i is the number of particles in the i th size interval; M_i is the mass associated with particles in the i th size interval; ρ is the density of the particles; and D_i is the average particle diameter in the i th interval. Photometer response for particles in the i th interval was estimated by multiplying the relative photometer response (Fig. 2) for D_i times N_i . The relative photometer response was determined by interpolation between the 100 individual calculations for particles between 0.1 and 1.0 μm in size. A best fit curve was taken from the data of Sinclair⁽¹⁾ to determine relative photometer response for particles larger than 1.0 μm .

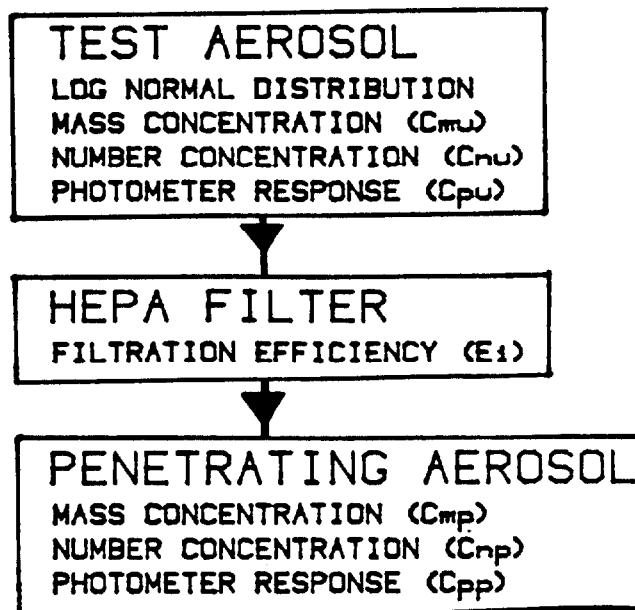


FIGURE 3
FLOW DIAGRAM FOR COMPUTER MODEL OF EFFICIENCY MEASUREMENTS

Aerosol mass and number concentrations, and photometer response was calculated for each size interval, and also summed over all size intervals, for the upstream aerosol. These same data were determined for the aerosol penetrating the HEPA filter (downstream aerosol) based on the number of particles in each size interval times a HEPA filter efficiency factor obtained from a filter efficiency curve (discussed below) for the average particle size in each size interval. These upstream and downstream data are then used to calculate overall filter efficiency based on aerosol mass, particle number, and photometer readings.

While Mie theory and physical measurement of the photometer optics provide a sound basis for defining relative photometer response, filter efficiency as a function of particle size varies between filters so some representative values had to be used. Four different filtration efficiency curves were considered. The first, shown in Fig. 4, is a second order exponential fit to data from an experimental measurement of DEHP penetration by Schuster and Osetek(6) in the particle size region of 0.1 to 0.3 μm . The high efficiency of HEPA filters have resulted in very little information on the penetration of micron size particles. The efficiency curve for particles larger than 0.35 μm was based on a smooth transition from the curve fitting the data to a penetration of essentially zero (3×10^{-10}) at 3.0 μm .

The QA test specifies the satisfactory performance of HEPA filters as an efficiency of at least 99.97 per cent for particles of 0.3 μm diameter. An important aspect of the QA test is the sensitivity to detection of a filter that just satisfies or just fails this minimal performance criteria. The evaluation of the QA test performed on a border line filter required the use of a hypothetical filtration efficiency curve. The shape of the curve shown in Fig. 4

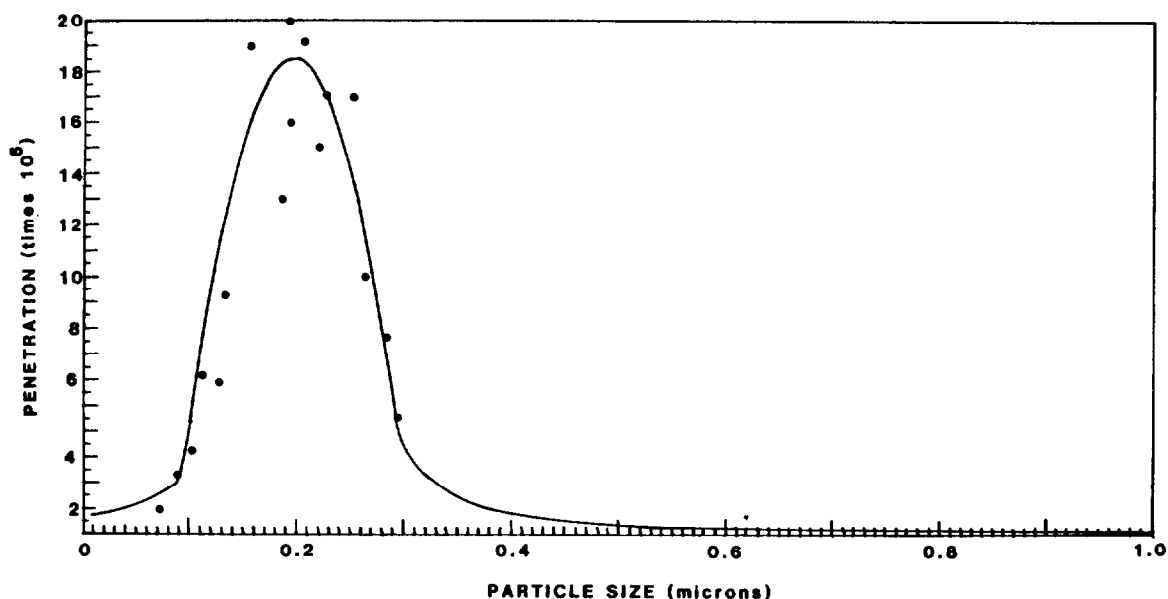


FIGURE 4
HEPA FILTER PENETRATION FOR DEHP PARTICLES (SCHUSTER AND OSETEK)(5)

was used to create the hypothetical curve. This efficiency curve was selected because the QA test is based on the assumption of a particle size that is most difficult to filter with a subsequent, easiest to measure lower efficiency. This type of efficiency curve (Figs. 4, 5, and 6) is consistent with the concept of several filtration mechanisms (diffusion, impaction, and interception). The hypothetical efficiency curve was created by displacing the particle size axes in Fig. 4 to set the maximum penetration at a particle size of $0.3 \mu\text{m}$. The amplitude of the curve in Fig. 4 was increased by multiplication by a constant factor to provide a penetration of 0.0003 for the maximum penetrating particle size of $0.3 \mu\text{m}$. The resulting curve is shown in Fig. 5. This curve has been created to match the single specified test condition in the QA test. Efficiency measurements that have been carried out to date(6,7) indicate the hypothetical low efficiency curve is not representative of high efficiency filters with respect to magnitude of penetration or particle size of maximum penetration. The shape of the curve is similar to most experimental filtration efficiency curves.

A filter efficiency curve from Dupoux and Briand(7) was also used, as this curve (Fig. 6) extends over a wider range of particle sizes. The maximum penetration observed by Dupoux and Briand (7) is about an order of magnitude higher than was observed by Schuster and Osetek.(6) For the reasons previously noted, the data of Fig. 6 was also adjusted (particle sizes increased by $0.15 \mu\text{m}$ and penetration multiplied by 2.6) to provide a maximum penetration of 0.0003 at a particle size of $0.3 \mu\text{m}$ for one set of efficiency calculations.

Filtration efficiency curves cannot be uniquely defined as they vary between filters and possibly by the method used to measure efficiency. However, for the purposes of this study, it is sufficient

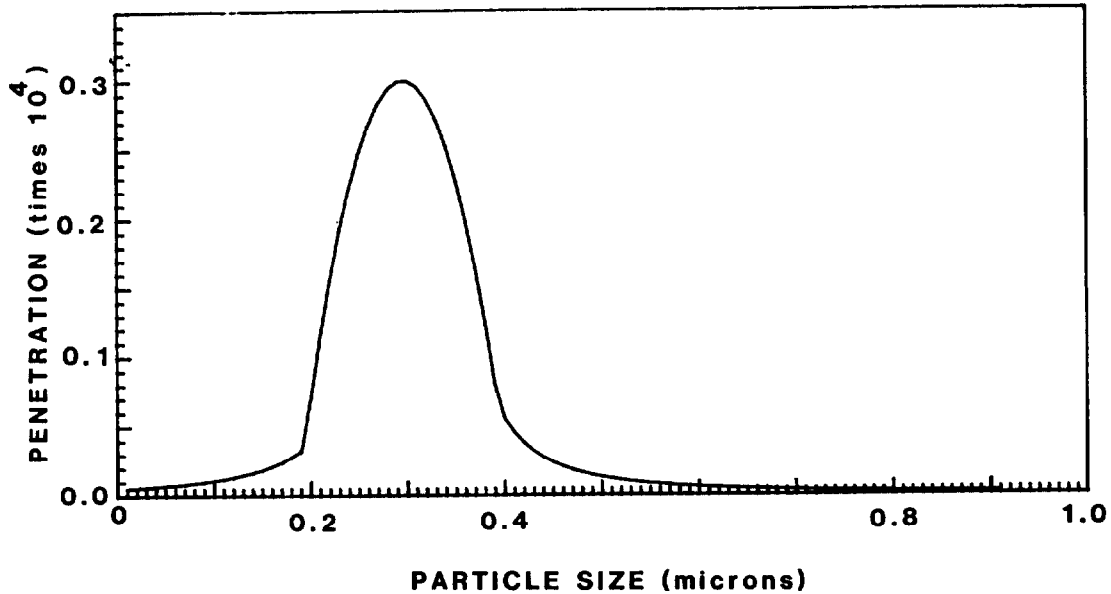


FIGURE 5
HEPA FILTER PENETRATION CURVE FROM SCHUSTER AND OSETEK TRANSPOSED TO
MAXIMUM PENETRATION OF 0.0003 AT 0.3 μm

that the efficiency curve has a particle size variation that is characteristic. Figures 4, 5, and 6 all are similar to most experimental efficiency measurements with some difference in magnitude and location of the peak. The monotonic response of the photometer shown in Fig. 2 indicates that location of the peak anywhere between 0.05 to 0.50 μm will have little effect on the calculations. The two experimental efficiency curves give overall efficiencies very similar to the results of experimental measurements of HEPA filters.

In all four cases, the filter efficiency calculation was checked by use of a computer generated challenge aerosol having (1) the same volume median diameter as the aerosol of interest, but a geometric standard deviation of 1.05 (approximately monodisperse) and (2) a count median diameter (cmd) that matches the size of maximum penetration for the efficiency curve also with a geometric standard deviation of 1.05. In all of these cases, all three efficiencies (mass, number, and photometer) were identical and matched the filtration efficiency curve values for the median diameters. The calculations for monodisperse aerosols confirmed the performance of the calculation procedure.

The efficiency curves used in this model may not be representative of the efficiency curves of filters having performance that is marginal or performance that would fail the QA test. The curves were obtained with filters that were significantly better than the acceptance criteria. It is possible that filters having efficiencies below 99.97 per cent will not have sharp peaks in their efficiency curves. However, at the present time there is no information on the performance characteristics of filters that do not meet the acceptance criteria. This indicates the need for a better definition of

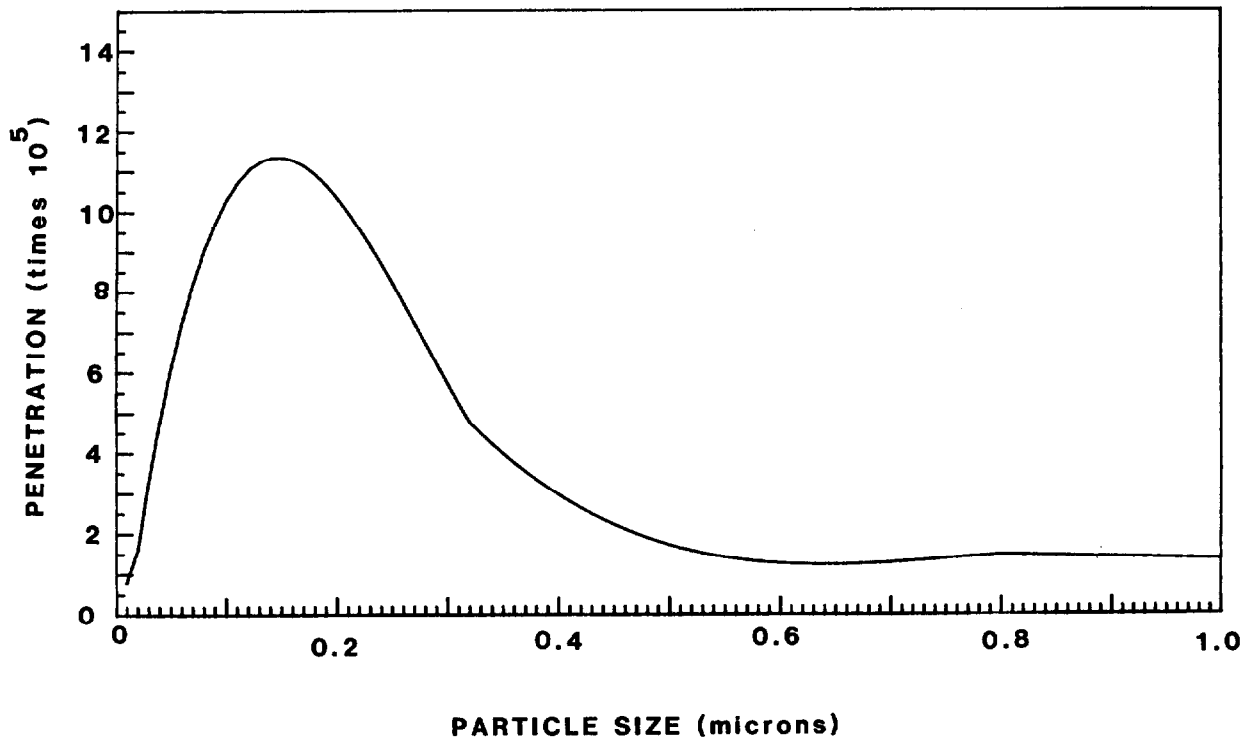


FIGURE 6

FILTER PENETRATION AS A FUNCTION OF PARTICLE SIZE (DUPOUX AND BRIAND)

filter efficiency as a function of particle size based on both experimental and theoretical work. If the efficiency curves vary significantly between acceptable and non-acceptable filters, then the type of efficiency measurement that is being made may also vary significantly. The results of this study should be representative for normal operating or acceptable filters.

V. Results

Filtration Efficiency Curve Varied for Three Experimentally Measured Aerosols

The efficiencies calculated for the three aerosols observed at the FTF are given in Table I. The reference column indicates if the filter efficiency model is based on the work of Schuster⁽⁶⁾ or Dupoux.⁽⁷⁾ The size at minimum efficiency column indicates if the filter efficiency model has been adjusted to produce minimum efficiency at 0.3 μm diameter, or if it is based on the experimental data which indicates the size of maximum penetration is 0.196 μm ⁽⁶⁾ or 0.146 μm .⁽⁷⁾

Except for one test, the difference between the per cent efficiency based on particle mass, particle number or photometer reading for the three aerosol sizes analyzed are all less than 0.01 per cent. This magnitude of difference would have a negligible effect on overall efficiencies specified to .01 per cent.

Table I. Filtration Efficiencies

Aerosol Characteristic			Filter Efficiency Curve			Calculated Overall Removal Efficiencies (%)			
Material	CMD1 (µm)	GSD2	Minimum Efficiency (%)	Size at Minimum Efficiency (µm)	Efficiency for 0.3 µm Diam. Particles	Reference	Particle Mass	Particle Number	Photometer Reading
DEHP	0.18	1.38	99.9982	0.196	99.9996	6	99.999	99.999	99.999
DEHP	0.18	1.38	99.9887	0.146	99.9943	7	99.992	99.990	99.995
DEHP	0.18	1.38	99.970	0.300	99.970	6	99.984	99.991	99.983
DEHP	0.18	1.38	99.970	0.300	99.970	7	99.980	99.987	99.977
DEHP	0.19	1.45	99.9982	0.196	99.9996	6	99.999	99.999	100.00
DEHP	0.19	1.45	99.9887	0.146	99.9943	7	99.994	99.991	99.996
DEHP	0.19	1.45	99.970	0.300	99.970	6	99.985	99.990	99.988
DEHP	0.19	1.45	99.970	0.300	99.970	7	99.979	99.985	99.981
DEHS	0.17	1.34	99.9982	0.196	99.9996	6	99.999	99.999	99.998
DEHS	0.17	1.34	99.9887	0.146	99.9943	7	99.991	99.990	99.990
DEHS	0.17	1.34	99.970	0.300	99.970	6	99.985	99.992	99.988
DEHS	0.17	1.34	99.970	0.300	99.970	7	99.981	99.988	99.982

1Count Median Diameter
2Geometric Standard Deviation

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These results also indicate that the QA test conducted with these aerosols does not provide a consistent or accurate measurement of the criterion specified for the QA test. The QA test is supposed to measure the filtration efficiency for 0.3 μm diameter particles. In general (75 per cent of the measurements) the photometer measured efficiency is greater than the actual filtration efficiency for 0.3 μm diameter particles.

Effect of Aerosol Parameter Variation on Efficiency Measurement

The correlation between efficiency measurement method and characteristics of the particle size distribution was investigated by calculating the filtration efficiency with respect to particle number, and photometer measurements for aerosols having volume median diameters up to 5 μm and geometric standard deviations up to 2.5.

These calculations were carried out for all four filter efficiency models. The models based on actual experimental efficiency measurements (Figs. 4 and 6) indicated no photometer response to the downstream aerosol when the challenge aerosol has a volume median diameter of greater than 1.0 μm . Measurements made under these conditions would provide little information on filter performance. The lower efficiency curves derived by adjustment of the two experimental efficiency curves to 99.97 per cent efficiency for 0.3 μm particles illustrate the effect particle size distribution can have on efficiency measurements. These results are shown in Figs. 7 and 8. The photometer measured efficiencies are lower than the number efficiencies (Photometer Eff - Number Eff ≤ 0) when the challenge aerosol has a small median diameter ($D \leq 0.5 \mu\text{m}$). This diameter increases as the geometric standard deviation increases. The photometer measured efficiency is greater than the number efficiency when the median diameter is greater than about 1 μm with the difference increasing as geometric standard deviation increases. The range between 0.5 and 1.0 μm is interesting as in place filter tests use aerosols in this size range. There appears to be some possibility for significant differences in this range. However, the reason for these differences is not apparent.

VI. Discussion

The calculated efficiencies in Table I indicate the photometer will provide a reasonable and often conservative measure of "true" filter performance for the FTF test aerosols. However, the measured efficiencies do not provide a good correlation with the criteria (efficiency at 0.3 μm) specified by the QA test. This is probably the result of the very sharp peak in the filtration efficiency curves (i.e., Fig. 5). Any polydispersity in the test aerosol or any difference in the mean diameter of a monodisperse aerosol from the diameter specified for the test (0.3 μm) can result in significant differences between the measured filtration efficiency and the actual efficiency for monodisperse 0.3 μm particles. Significant in this case means differences that could result in errors in acceptance or rejection of filters.

The results shown in Figs. 7 and 8 indicate that the photometer measured efficiencies will be lower than the actual number efficiencies when the particles are small, where the relative response of the

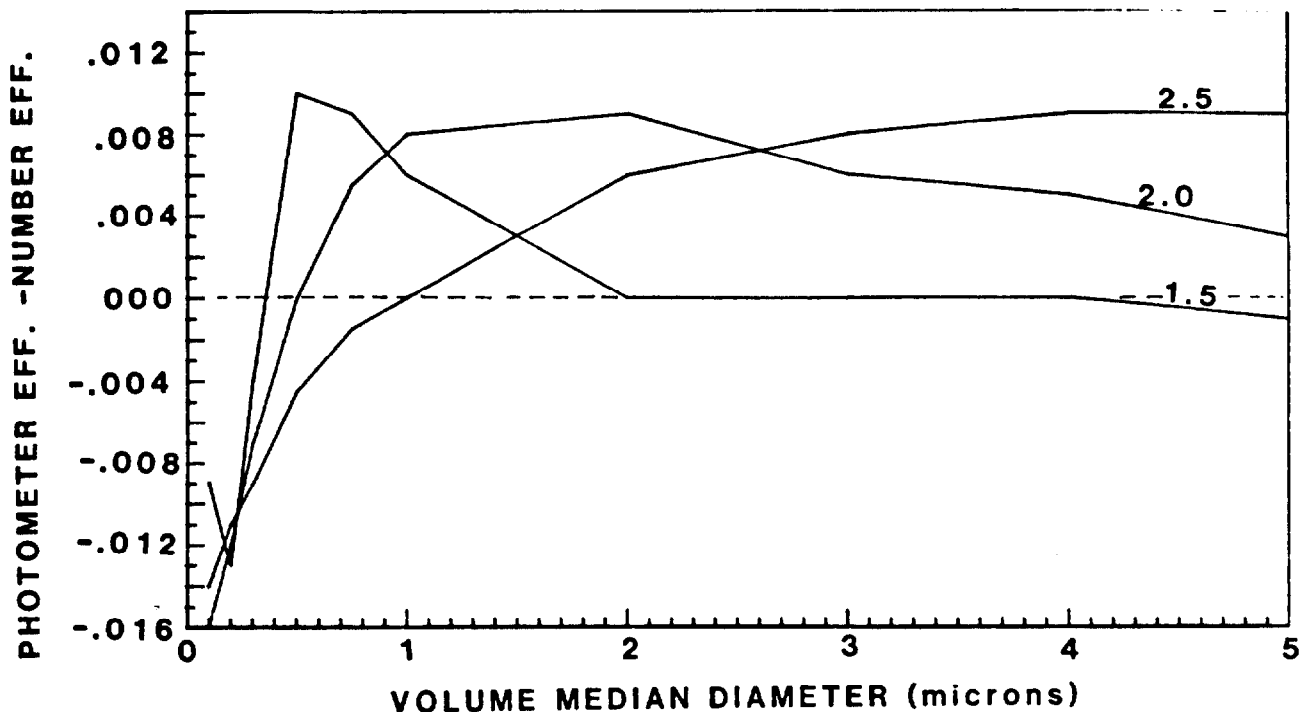


FIGURE 7

DIFFERENCE BETWEEN EFFICIENCY CALCULATED FOR THE PHOTOMETER READING AND CALCULATED NUMBER EFFICIENCY FOR SCHUSTER AND OSETEK EFFICIENCY CURVE ADJUSTED TO 99.97 PER CENT EFFICIENCY AT $0.3 \mu\text{m}$, GEOMETRIC STANDARD DEVIATION EQUAL = 1.5, 2.0, and 2.5.

photometer is low. The use of a test aerosol having larger particles will result in an overestimate of the number efficiency. There is, of course, less difference between efficiency with respect to mass and the photometer measured efficiency, since the larger particles make the most significant contribution to the airborne mass and the relative photometer response is much higher for the larger particles.

The most significant difference between the test as it is often conducted with a polydisperse aerosol having a median diameter of less than $0.3 \mu\text{m}$, is that the test does not measure the performance specified by the test. The filters that leave the filter test facilities carry a stamp saying they will remove at least 99.97 per cent of the $0.3 \mu\text{m}$ particles. However, these results indicate that slight variations in the challenge aerosol result in almost no information about filter performance with respect to $0.3 \mu\text{m}$ diameter particles. These calculations also indicate that specification of filter performance with respect to a single particle size provides extremely limited information. The results provided by the QA test are usually a conservative or an underestimate of filter performance with respect to the challenge that will be encountered in the field. The test results with the polydisperse aerosol are actually more representative of filter performance than the efficiency measured for $0.3 \mu\text{m}$ diameter particles.

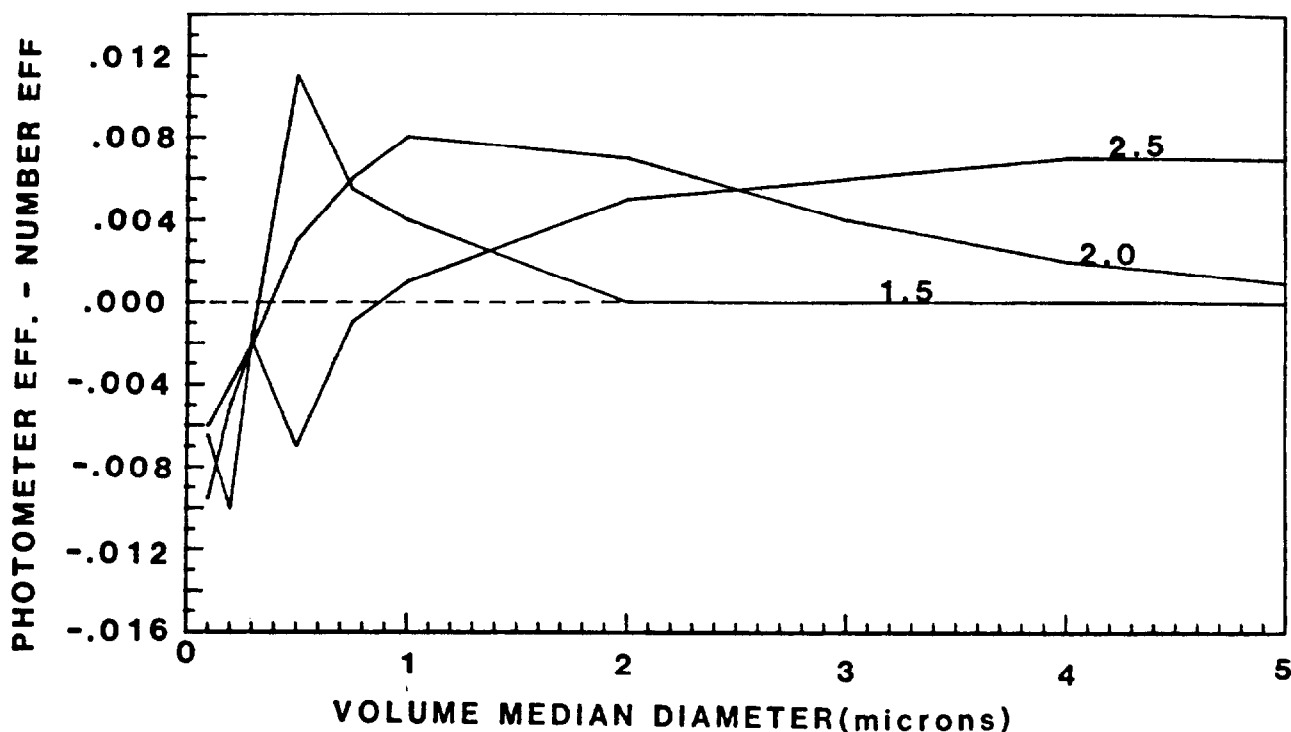


FIGURE 8

DIFFERENCE BETWEEN EFFICIENCY MEASURED BY PHOTOMETER AND NUMBER EFFICIENCY FOR DUPOUX AND BRIAND EFFICIENCY CURVE ADJUSTED TO 99.97 PER CENT EFFICIENCY AT $0.3 \mu\text{m}$, GEOMETRIC STANDARD DEVIATION = 1.5, 2.0, and 2.5.

The differences calculated and discussed between the various efficiencies are quite small, but it is important to remember that a penetration of .0025 is acceptable while a penetration of .0035 means the filter is rejected.

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6. B. G. Schuster and D. J. Osetek, "Multiple HEPA Filter Test Methods, March 21, 1975 - December 31, 1975," LA-6443-PR (August 1976).
7. J. Dupoux and A. Briand, "Air Filtration as a Function of Particle Size and Velocity," Water, Air, and Soil Pollution, 3, p. 537 (1974).

DISCUSSION

BERGMAN: Did you correct for multiple scattering in your photometers and did you make response vs. size calculations? When Mie theory results are modified for multiple scattering there is a very significant correction. You can get a rough feel for the magnitude of the effect experimentally if you measure specific response, corrected for concentration. Did you do something like this and are you aware of concentration effects?

TILLERY: We used Mie theory to do the calculations, but the focal point, in this case, is very small, so we did not make corrections for multiple scattering. I don't think it would be very significant in this type of illumination.

DYMENT: Is it correct to interpret your results as justifying a preference for a test using a polydisperse aerosol rather than a monodisperse aerosol? If so, what would be optimum mass median diameter and geometric standard deviation?

TILLERY: It is difficult to generalize from four efficiency trials but the general indications are that you are going to provide much more information about the performance of a filter when you challenge it with a range of particle sizes. One of the principal reasons is because of the peak in the efficiency curves, that is, if the peak is a valid measurement of the efficiency and the slope is very steep from both sides of the peak. Therefore, if your choice of particle is away from that peak, you are going to get a measurement that is markedly different from the overall performance of the filter. The efficiency curve is for a normally-performing filter, whereas what we are looking for is a filter that does not satisfy the performance criteria. We really don't know what the efficiency curve looks like for the type of filter that we are trying to match, i.e., the ones that do not satisfy the performance criterion. My feeling is that I agree with you but it is difficult to generalize from the measurements that have been made so far.

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PERFORMANCE OF 1000- AND 1800- CFM HEPA FILTERS ON LONG EXPOSURE TO LOW ATMOSPHERIC DUST LOADINGS, III

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Abstract

Comparative tests have been in progress since December 1977 to determine the performance of a number of standard (1000 CFM capacity) and high volume (2000 CFM) HEPA filters when exposed to ambient dust loadings. All standard filters have been tested at their rated capacity of 1000 CFM whereas the high volume filters have been tested at both 1800 and 1000 CFM. Initial results reported at the 15th and 16th Nuclear Air Cleaning Conferences indicated that an increase in service life of 1.6 times could be anticipated for the 1800 CFM rated filters when operated at 1000 CFM. This increase in service life is only half the theoretical increase. Additional tests have been conducted to determine the effect of prefilters on the service life of 1800 CFM rated filters when operated at 1800 and 1000 CFM. The results of tests with prefilters are the principal subject of this paper.

I. Introduction

This is the third in a series of reports^(1,2) on the comparative service life of HEPA filters constructed in conformance with US standard MIL-F-51068⁽³⁾ and those of more recent continental European design. Although the European design filters occupy no more volume than the US design filters, they contain almost double the area of filter paper. The increase in filter area within the same space is achieved by folding the paper into closer pleats and assembling unitized pleated paper packs into holding frames in a different configuration. Because of the presence of larger amounts of filter paper, the European design filters are able to handle 80 to 100% more air for the same friction loss when the filters are newly installed.

Two questions initiated this investigation in 1977: 1) how would the performance of European design filters compare with that of US design filters when subjected to the same aerosol at the rated flow of each and 2) how would the European design filters perform when downrated and used as one-to-one replacements for US design filters? Theoretically, a filter with twice as much of the same paper should give four times the service life when operated at half capacity, i.e., the airflow resistance should be reduced to one-half when the filtration velocity is halved and by another half when the dust deposited on each square foot of filter paper is halved. Our prior studies^(1,2) have shown that the theoretical increase in service life is not achieved. In fact, the service life increase obtained by downrating the European design filters was only half the theoretical prediction. On examination it was found that the air passages between the close pleats were being bridged by what appeared to be coarse fibrous material, whereas the widely spaced pleats of the US design filters appear able to store more of the coarse fibrous material in the air passages before the bridging process began. But inevitably even the air passages of the US design filters bridged, resulting in an enormous increase in the

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rate of pressure rise. Analysis of filter behavior suggested that the use of a prefilter to remove the coarse, fibrous dust might result in a worthwhile increase in the service life of a downstream European design HEPA filter, whether operated at rated capacity or downrated to run at 1000 CFM. Filter tests of this kind were initiated in the summer of 1980 and are continuing to the present. This report will be concerned primarily with HEPA filter performance while using prefilters.

Prefilters

The prefilters that were used were of a single type: 24 x 24 in. frames containing 2-in. deep pads of graded polyester fibers. The upstream side contained 40 denier (66 μ m) fibers and the downstream side, 4 denier (21 μ m) fibers. The filters were manufactured by TRI-DIM Filter Corp.⁺ to fit our filter frames. Clean resistance at 1000 CFM (250 fpm superficial face velocity) was 0.1 in. w.g.; at 1800 CFM (450 fpm superficial face velocity, the clean resistance was 0.4 in. w.g. (Figure 1).

When operated at 1800 CFM, after an initial rapid rise of 0.1 in. w.g., airflow resistance of the prefilter increased monotonically with time for 116 days and reached a resistance of 1.34 in. w.g. before being replaced with a second filter of identical construction. The second 1800 CFM prefilter was in service for 243 days, increasing in resistance only 0.3 in. w.g. during that period. The reason for the very different resistance rise history of the two prefilters is not clear. It is believed that the second one became damaged after a period and shed its dust load onto the HEPA filter behind it because the HEPA filter began to rise at a much more rapid rate than it had done with the first prefilter in place. It is possible that winter precipitation penetrated the protective louvers and did the damage because the resistance of the prefilter operated at 1000 CFM rose at a steady but lesser rate just as did the first 1800 CFM prefilter. At about the time the resistance of the second 1800 CFM prefilter ceased increasing, the 1000 CFM prefilter showed a sudden drop in resistance. This occurred for both, about mid-December, 1980. The 1000 CFM prefilter maintained a low level of filter resistance through the winter and spring and during late spring the resistance rose rapidly to approximately 3.5 in. w.g. at which point, after 428 days of service, it was changed.

HEPA Filters

Although the number of trials with and without prefilters is limited, it is possible to come to some tentative conclusions regarding the effect on HEPA filter service life of using one type of low resistance fibrous prefilter. Figure 2 shows the resistance of European design 1800 CFM-rated filters (Delbag) when operated with and without the prefilters described in the previous section. When operated without a prefilter, a resistance of 3.0 in. w.g. was reached when 0.84 kg. of unfiltered atmospheric dust reached the HEPA filter and a resistance of 5.0 in. w.g. was reached by the time 1.02 kg. of dust had entered the filter. The HEPA filter with the prefilter operated for the first 116 days (approximately 0.5 kg. dust) with no

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significant difference in resistance buildup relative to the HEPA filter without a prefilter. After the replacement of the first prefilter with another of like construction, the resistance curves of the two HEPA filters began to diverge. At 3.0 in. w.g., the HEPA filter-prefilter combination had accumulated 0.94 kg. dust vs. 0.84 for the HEPA without prefilter and at 5 in. w.g. The HEPA-prefilter combination had accumulated 1.07 kg. dust vs. 1.02 kg. for the HEPA without prefilter.

Figure 3 shows the effect of operating a downrated (1000 CFM) European design HEPA filter (Poelman) with and without a prefilter. After 358 days of operation and the intake of 0.7 kg. of atmospheric dust, little difference in resistance could be observed between the HEPA filters with and without a prefilter. It is possible that neither filter had yet accumulated sufficient dust to enter a more rapid resistance rise regime.

A number of years ago many US nuclear installations made a conscious decision to eliminate the use of prefilters ahead of HEPA filters because in their experience only minor increases in HEPA service life were observed and the extension in HEPA filter life was more than offset by the labor cost of changing prefilters. There is nothing in the service life data accumulated so far in our tests that refutes the earlier decision regarding the non-use of prefilters. It is necessary to keep in mind, nevertheless, that (1) prefilters that differ in construction from the one used in our trials may give different, and more favorable, results with respect to prefilter usage and (2) there is considerable variability in our own results for the same HEPA filter when tested at different times, as demonstrated in Figure 4. This makes it dubious that firm conclusions should be drawn from the few trials that we have conducted.

II. Summary

On the assumption that it will prove useful to summarize all our data since 1977 in one place, Figure 5 and Table 1 have been included in this report, even though much of the information has already appeared in one or another of the two previous reports. Because DOE funding for this program ceased almost two years ago, it is doubtful that this series will continue.

III. References

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2. First, M.W. and Rudnick, S.N., "Performance of 1,000 and 1,800 CFM HEPA filters on long exposure to low atmospheric dust loadings II" Proceedings of the 16th DOE Nuclear Air Cleaning Conference, San Diego, California; USDOE Report CONF-801038; 1; pp 682-696; (2/81)
3. U.S. Military Specification MIL-F-51068, Filter, Particulate, High-Efficiency, Fire-Resistant, Edgewood Arsenal, MD.

Table I. Summary of Results

HEPA Filter Brand	Air Flow Rated Capacity	(CFM) Test Rate	Start up Date	Initial Pressure Drop (in w.g.) HEPA Filt.	Time for HEPA to Reach 3 in. w.g. (Days)	Dust Weight (Kg.) at Stated Resistance						Shut Down Date	Total Run Time (Days)
American Air Filter	2000	2030	11 Apr 79	0.84	262	1.6	1.73	1.75	-	-	-	9 Jan 80	273
America Design	a	1000	29 Dec 77	1.10	281	0.53	0.66	0.71	-	-	-	3 Nov 78	309
America Design	a	1000	17 Nov 78	1.10	276	0.69	0.86	0.91	-	-	-	24 Sep 79	301
Delbag	b	1770	3 Jun 80	1.13	227	0.64	0.84	0.97	1.02	1.02	-	7 Apr 81	309
Delbag + Prefilter	b	1770	3 Jun 80	1.17	270	0.72	0.94	1.02	1.07	1.10	-	14 July 82	401
Luwa		1770	29 Dec 77	0.49	497	1.10	1.18	1.24	1.29	-	-	10 Jul 79	534
Luwa		1770	16 Aug 79	0.52	784	1.56	1.65	1.71	-	-	-	8 Dec 81	847
Luwa		1770	29 Dec 77	0.82	281	1.00	1.14	1.16	1.17	-	-	17 Oct 78	289
Luwa		1170	17 Nov 78	0.92	323	1.56	1.75	1.84	1.87	1.90	-	14 Nov 79	347
MSA		2000	28 Aug 79	0.53	696	1.49	1.54	1.60	1.65	-	-	23 Nov 81	820
MSA		2000	20 Aug 79	1.15	308	1.41	1.80	1.82	1.83	1.86	-	10 Jul 80	319
Poelman		1770	10 Jan 79	0.50	955	2.09	2.30	2.42	2.46	-	-	23 Feb 82	1142
Poelman		1770	4 Jun 80	0.45	-	-	-	-	-	-	-	8 Sep 82	827
Poelman + Prefilter	c	1770	4 Jun 80	0.51	-	-	-	-	-	-	-	8 Sep 82	525
Poelman		1770	10 Jan 79	0.77	223	1.00	1.29	1.49	1.62	1.70	-	9 Nov 79	304
Vokes		2000	16 Jun 82	0.53	-	-	-	-	-	-	-	8 Sep 82	84
Vokes		2000	2 Jun 82	0.92	-	-	-	-	-	-	-	8 Sep 82	99

a. Luwa type paper

b. Prefilter changed after 116 days and after 241 days

c. Prefilter changed after 428 days

Note: Prefilters were changed when test flow rate through filter system could not be maintained.

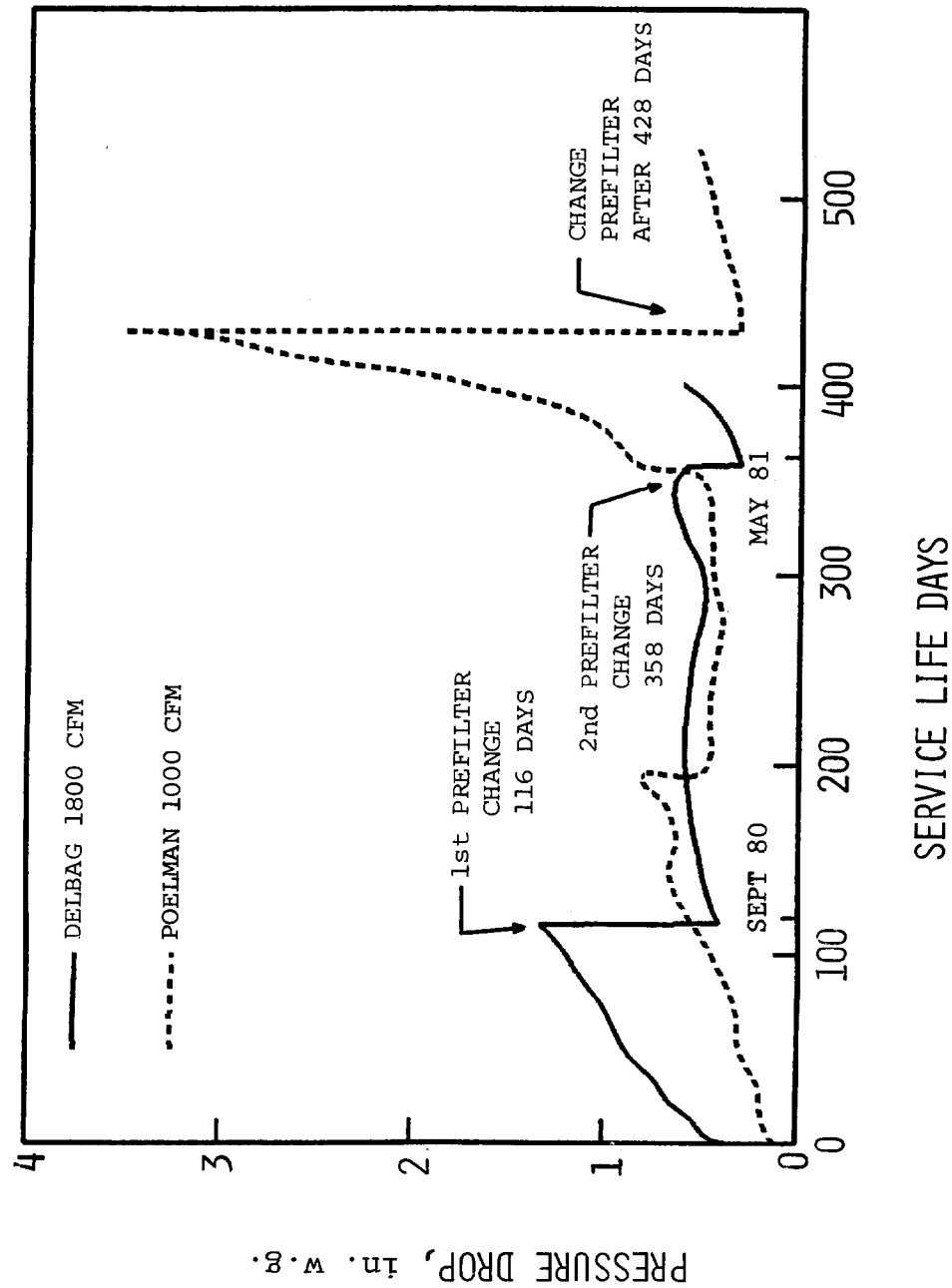


FIGURE 1: PREFILTERS: AIRFLOW RESISTANCE VARIATION WITH SERVICE TIME

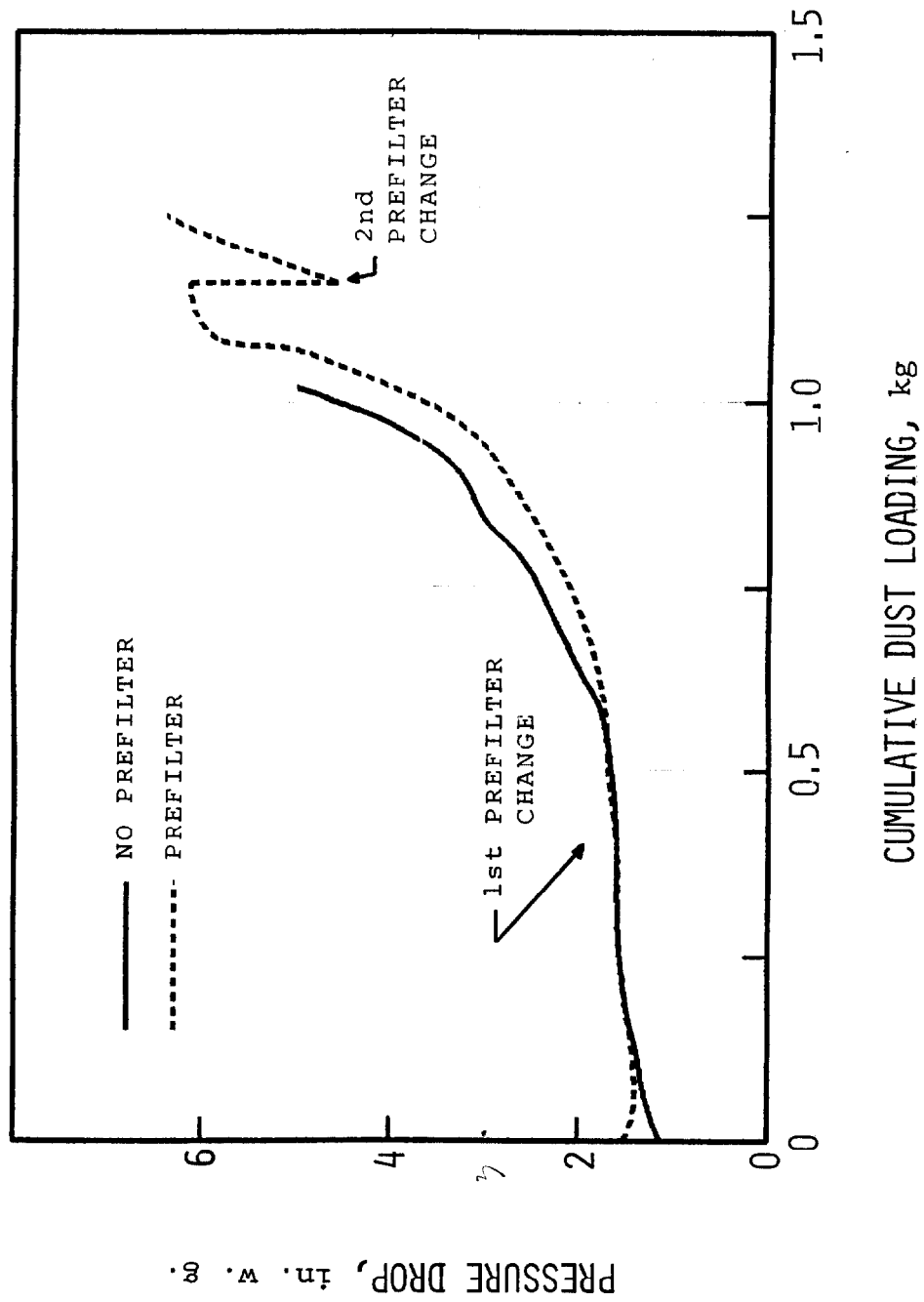


FIGURE 2: DELBAG 1800 CFM PERFORMANCE CURVE

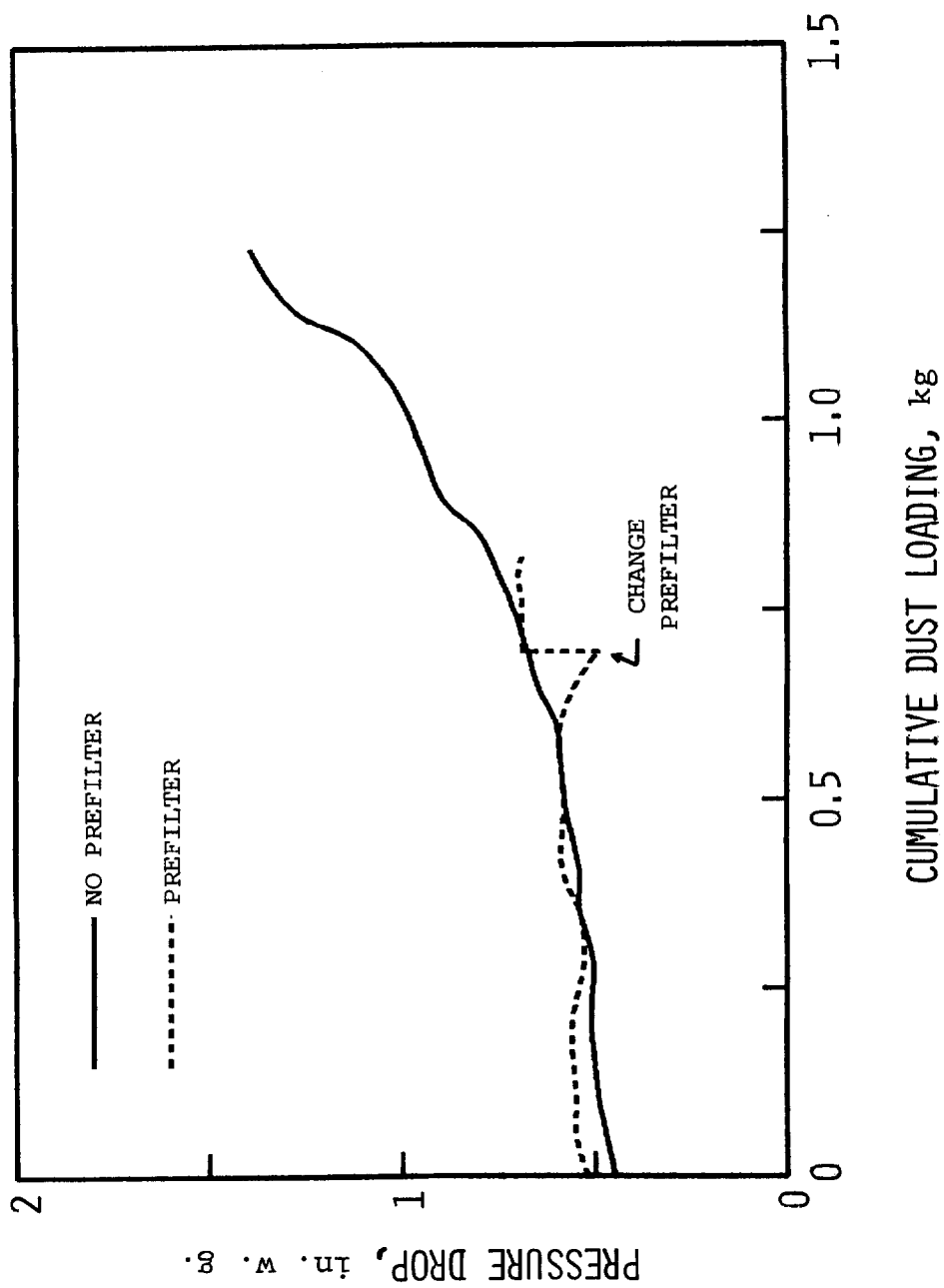


FIGURE 3: POELMAN 1000 CFM PERFORMANCE CURVE

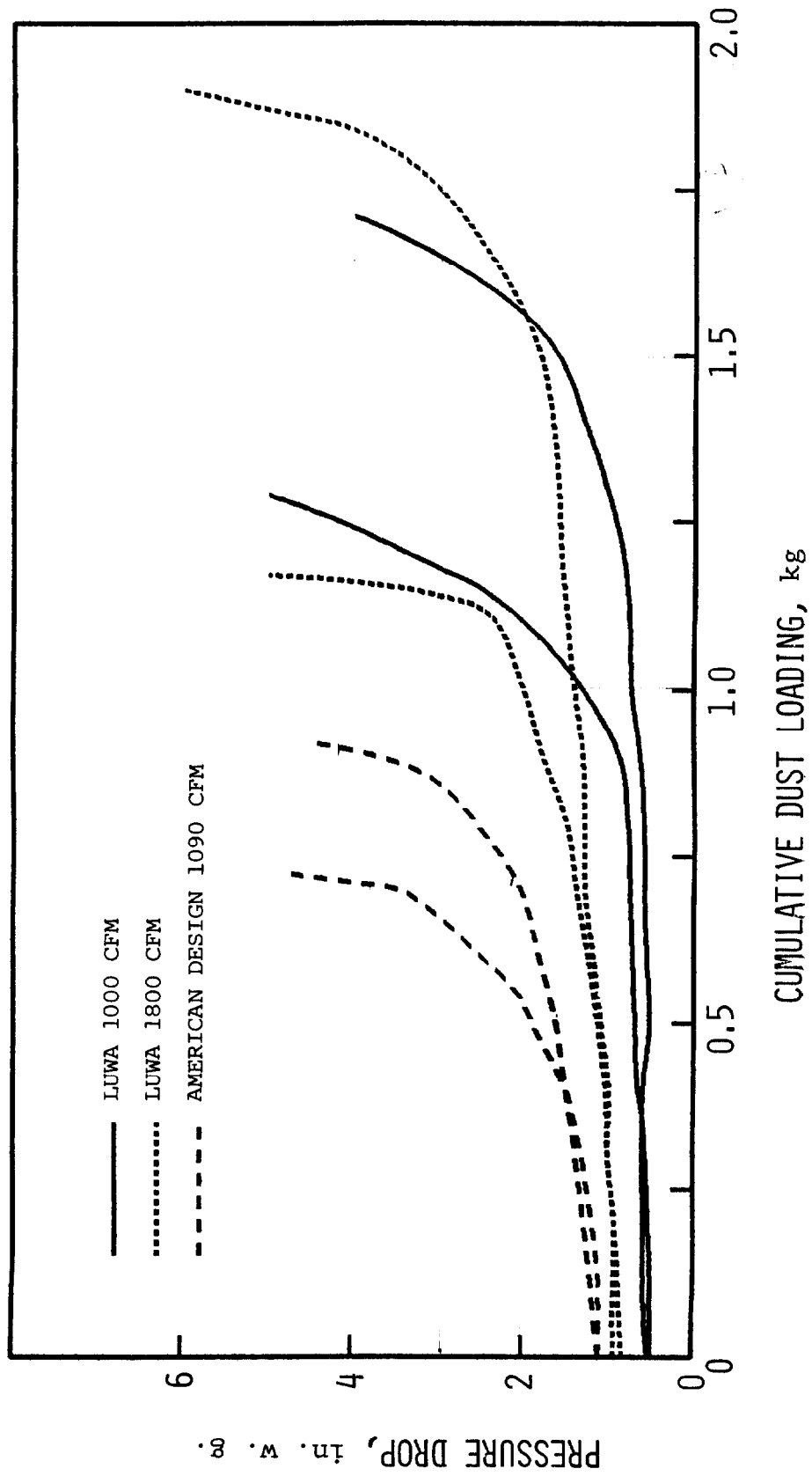


FIGURE 4: REPLICATE PERFORMANCE CURVES

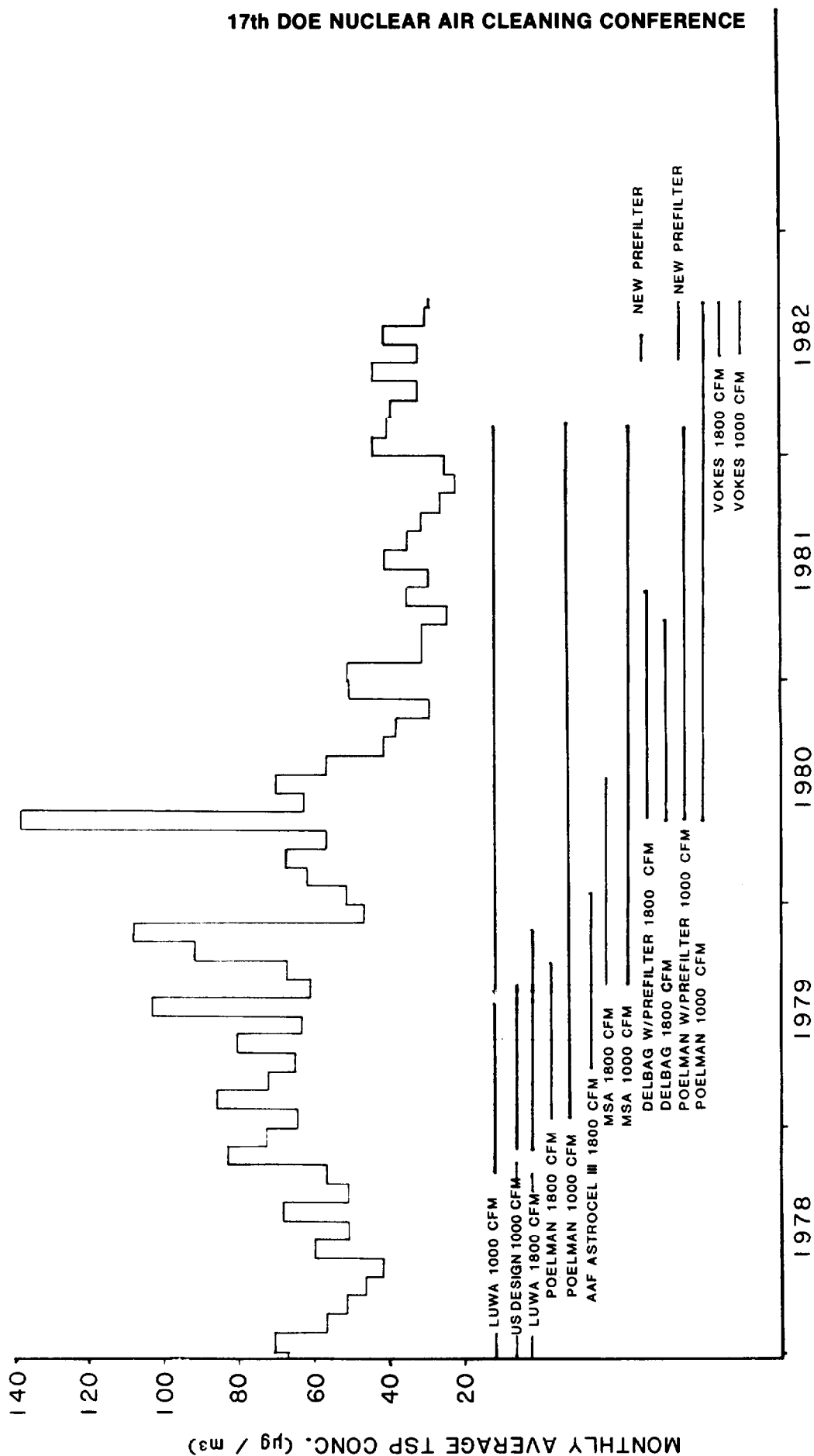


FIGURE 5: CHRONOLOGY OF TSP CONCENTRATION and HEPA FILTER TESTS

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DISCUSSION

BERGMAN: Did you measure the efficiency of the prefilters? The efficiency of prefilters is the key parameter that determines the extension of HEPA service life. In our studies, we have shown very large extensions of HEPA service life. We had an evaluation at Rocky Flats where the prefilter extended the HEPA filter service life by over 40 times. In another application, we were able to extend the HEPA filter life during fire and smoke conditions from 5 minutes to over 1 hour and the HEPA filter was still not plugged. The use of high efficiency, high loading prefilters provides a significant extension of HEPA filter life in nuclear applications. The primary argument against the use of prefilters is the cost of changing prefilters. This disadvantage is readily overcome by designing a method for changing prefilters in ventilation systems without shutting the system down or entering large plenum chambers.

FIRST: Prefilter efficiency was not measured, an austerity measure we had to adopt in the absence of financial support for the program. With regard to your second point, if we accept your assumptions, which I have no problem with, the conclusions which you have reached seem reasonable. Please keep in mind that the primary purpose for our prefilters (which are far from 95% efficient for atmospheric dust) was to remove coarse fibers and coarse dust which we had assumed were plugging the interstices of the pleats. From our data, this proved to be incorrect and the resistance rise of the HEPA filters is exclusively the fine particles which penetrated the particular prefilter that we were using.

BERGMAN: The reason I am concerned about an assessment of the use of prefilters in the nuclear industry for protection of HEPA filters is that from almost every study I have seen, the conclusion is that prefilters have a pronounced effect on prolonging HEPA service life.

DYMENT: In the light of the test results with prefilters, can you suggest an explanation for the apparent failure of the high flow HEPA unit to utilize all of the available area of paper? The suggestion that coarse dust was bridging interstices presumably is not now supported by the facts.

FIRST: I regret that I do not have a satisfactory explanation for you. I have thought that the answer may lie in the narrowing of the small air spaces between the pleats as the dust builds up on either side of the passage, but I have no data to back this up.

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PORTABLE FILTER TESTING INSTRUMENTATION USED FOR IN-PLACE LEAK TESTING OF LARGE AIR FILTERS UP TO $38 \text{ m}^3 \text{ sec}^{-1}$ (80,000 CFM)

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ABSTRACT

The demand for testing large aerosol and gas filter systems in nuclear power plants resulted in the design of a new generation of NUCON* F-1000 field test instrumentation. This consists of a thermal aerosol generator, and an aerosol detector used for testing HEPA filter banks, as well as a halide gas generator and a pair of halide detectors, for testing adsorber banks.

The DG-F Thermal Aerosol Generator is electrically heated and weighs only 22 kg (50 lbs). It transforms up to 130 gram per minute of liquid DOP (Diocetyl-Phthalate) into 0.7 micron average size aerosol. The generator can be modified for use with other comparable test agents as well.

In the DD-SA forward light-scattering aerosol detector, the traditionally used photomultiplier tube is replaced by a silicon photodiode - operational amplifier combination. This increases the long term stability and reliability in spite of rough handling in transportation.

The HG halide generator generates halide vapor by evaporating R-11, R-12 or other common halides with excellent concentration control.

The already proven advantages of constant readout halide detectors versus gas chromatographs for adsorber leak tests are enhanced with the new HD-SA detectors.

The servo-stabilization of gas sampling velocity regardless of back pressure, the direct readout of gas concentration with 1Vppb precision, the built in timer for sequential readouts with 1 second precision enables testing personnel to perform reliable and reproducible leak tests in minimum time.

Both the generators and detectors were designed to meet the testing requirements of ANSI N510-80 and similar standards for detecting in-place leak rates of less than 0.01% and have been field tested extensively to assure reliable operation.

I. INTRODUCTION

While both aerosol and halide gas detectors have existed in the past, these were mainly suitable for testing in a single location. The use of non-field type laboratory instruments is unsuitable for typical multi location testing. The NUCON field test department initially used such instrumentation and the damage rate occurring during transportation, even in well protected packaging, was very high. Even when obvious physical damage was prevented, these frequent adjustments and recalibration requirements caused unjustifiable testing delays. These initial problems led to the development of light weight air transportable test instruments which while maintaining or exceeding the sensitivity and repeatability of laboratory test instruments are eminently suitable for field testing due to their ruggedness and stability.

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The instrumentation was developed in conjunction with the personnel who actually perform the testing and are aware of the field replacements. The development started ten years ago (1972), and numerous refinements and state of the art technology was incorporated to achieve the utmost in reliability of both halide and aerosol leak testing of both nuclear and other critical air filtering applications.

II. AEROSOL DETECTOR

One of the classical methods of aerosol detection is the use of the forward light-scattering resulting from aerosol particles, utilizing a light source to produce a cone of light with a dark center and a photomultiplier (P.M.) tube placed in the dark area. The light-scattering caused by the particles, increases the current in the P.M. tube proportional to the concentration of aerosols, (See Fig. 1).

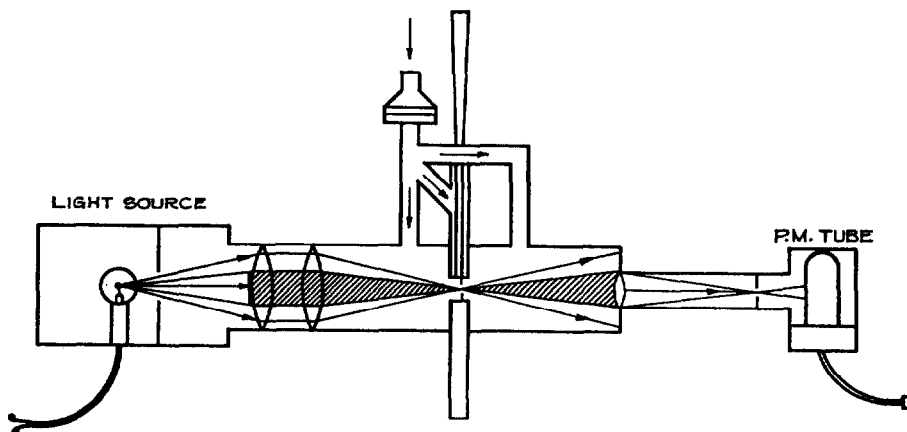


Fig. 1.

DETECTOR OPTICS CHAMBER

The aerosol detector designed specifically for field use by NUCON is based on this classic forward light-scattering principle, but uses carefully selected and matched components to assure reliable measurements at high sensitivity.

The Light Detector

P.M. tubes require high voltage and suffer from numerous other disadvantages, therefore, a silicon photodiode - operational amplifier (OP-AMP) combination was selected for the scattered light detection. The advantages of this unit are:

- a) The photodiode is less sensitive to vibration and shock than a P.M. tube.
- b) It operates at low voltages.
- c) It is exceptionally stable (drift often less than $\pm 2\%$ /year).
- d) It is not harmed by direct sunlight, therefore, does not require special precautions during assembly or servicing even while in operation.
- e) The photodiode has no "memory" if overexposed by light, thus fast changes in light quantity can be measured without error, while P.M. tubes exhibit "fatigue" and up to 40% short term sensitivity variations.
- f) The spectral response of the photodiode is wider than the P.M. tube, while the sensitivity is comparable to that of the P.M. tube, as shown on Fig. 2.

The Light Source

A 30 watt quartz halogen lamp was selected because it is reliable, stable and its spectral emission matches well the response of the photodiode. The spectral output of this bulb is also shown on Fig. 2.

The filament has constant intensity during its 400-2000 hour life due to tungsten recovery by the hot filament from the overheated quartz envelope.

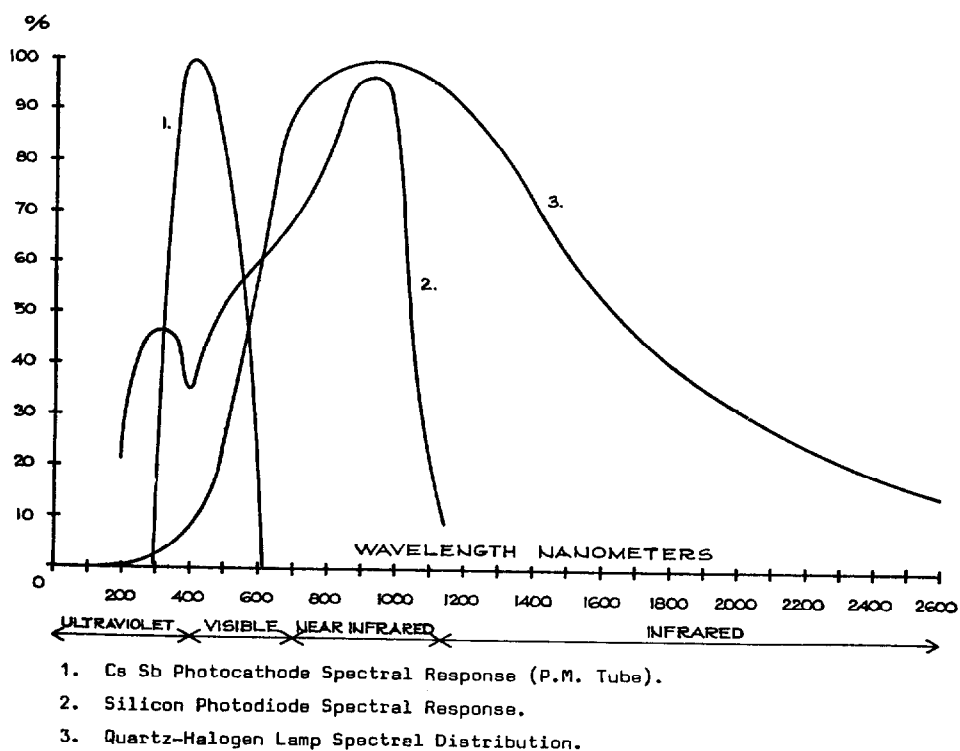


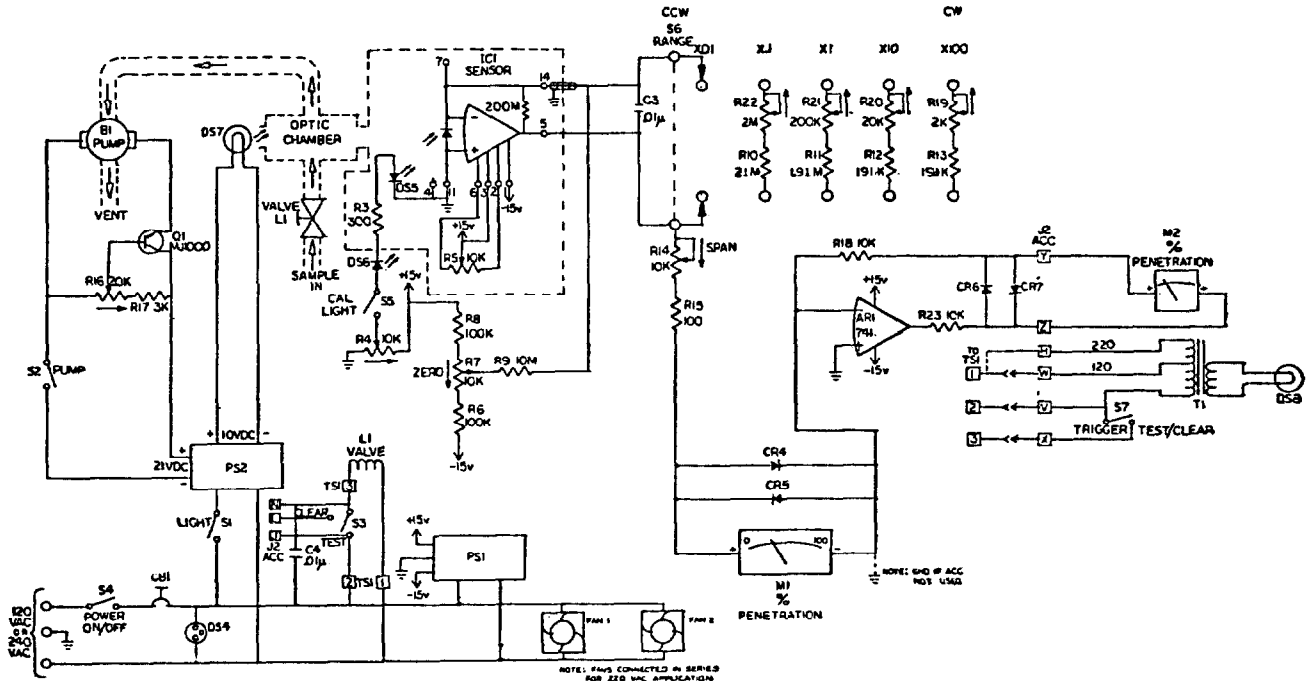
Fig. 2.

The Amplifier

The gain control of the OP-AMP is achieved by feedback resistors in steps of ten. The range switch covers four orders of magnitude and can be shifted both up or down another order of magnitude by the use of the span adjustment. The output current of the OP-AMP, which is proportional to the scattered light and the aerosol content, is displayed by a back-to-back diode protected microammeter.

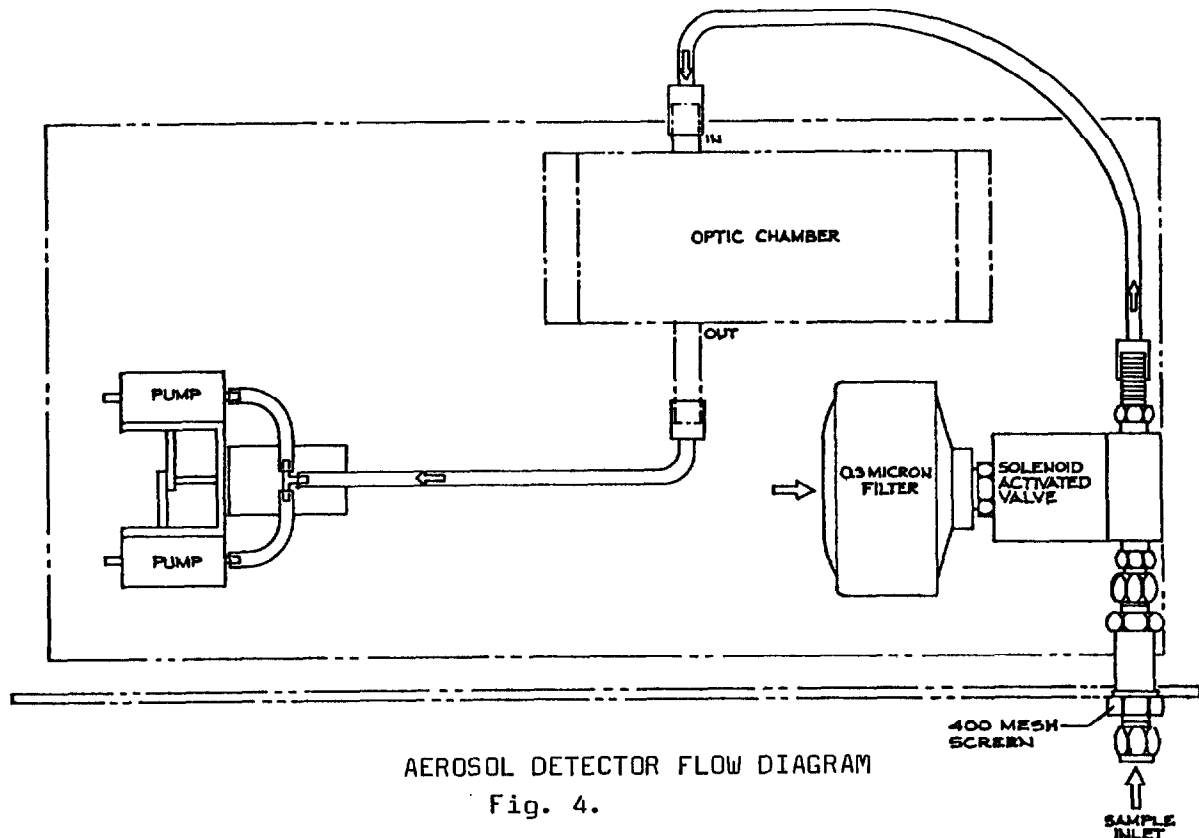
Compensation for straylight effects, resulting from the internal reflections in the optical system is achieved through the use of a zero bias potentiometer.

The linearity check of the detector and the field calibration, is by a light emitting diode powered by a stabilized power supply, and adjusted by a ten turn potentiometer. The schematic diagram is shown on Fig. 3.



AEROSOL DETECTOR SCHEMATIC

Fig. 3.



AEROSOL DETECTOR FLOW DIAGRAM

Fig. 4.

The Sampling Pump

A double piston diaphragm pump coupled to a brushless DC motor provides the air flow through the optical chamber as shown on Fig. 4. The motor speed is adjustable to accommodate the isokinetic sampling requirements. The lifetime of the DC motor is estimated at 10,000 hours while the pump service life is rated at 5,000 hours.

The sample inlet line is protected by a 400 mesh cartridge filter that prevents the entry of 38 micron and larger particles. The clear air intake is protected by a 99.97% efficiency (for 0.3 micron particle) gas mask style filter. A solenoid valve switches from "clear" to "sample" air. A hand held probe to scan for leaks is an optional detachable part of the detector system.

The Sensitivity of the Detector

Experimental gravimetric measurements of air samples on the input of the detector resulted in the following

TABLE I

Aerosol concentration providing 100% deflection

Span control zero (800) $\mu\text{g l}^{-1}$ (extrapolated)	Span control max 80 $\mu\text{g l}^{-1}$	Range switch position
80 "	8 "	100.0
8 "	0.8 "	10.0
0.8 "	0.08 "	1.0
0.08 "	0.008 "	0.1
		0.01

The mass of a one micron diameter particle of DOP is 5.23×10^{-13} g. The value $0.008 \mu\text{g l}^{-1}$ represents a concentration of 15.3 one micron diameter particles per cm^3 . In other words, the detector is able to sense a single particle of $1 \mu\text{m}$ diameter.

Physical Parameters

The entire instrument is housed within a portable metal case with dimensions of .13 m x .46 x .30 m, and weighs 10 Kg.

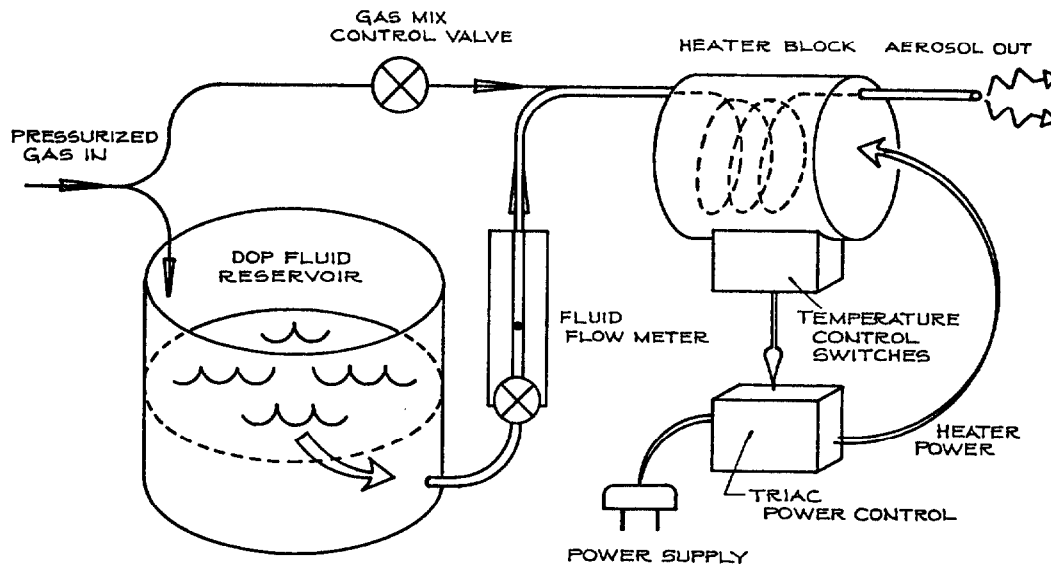
III. AEROSOL GENERATOR

The challenge agent Dioctyl Phthalate (DOP), used with the NUCON detection system, is a clear, viscous liquid utilized extensively in industry as a plasticizer. When heated past its boiling point of 385°C , and subsequently allowed to condense, an aerosol of less than 1.0 micrometer mean particle diameter is normally produced. The resulting aerosol has not only a desirable particle distribution, but also maintains its structure well, as a function of time due to DOP's high molecular weight, and low vapor pressure.

The NUCON thermal DOP aerosol generator is structured as (Fig. 5) illustrates. The liquid DOP contained within the generator's 1.4 liter holding reservoir, is forced by pressure through a flow control meter to an electrically heated block. Before entering the thermally regulated block, a flow of gas (usually less than 10 liters min^{-1}) is mixed with the liquid to aid in the manageability of the resulting aerosol.

Early attempts to generate sufficient volumes of DOP aerosol to test some of the larger filter systems (greater than 19 $\text{m}^3 \text{sec}^{-1}$, 40,000 cfm) proved very difficult, due in part, to the following problems:

- a) The energy requirements to vaporize the necessary volumes of liquid DOP are quite high. Generators had either too little power input, and/or did not possess the necessary heat transfer characteristics to accomplish the task for the period of time necessary to conduct a test.
- b) Generators that did have the needed capacity were exceedingly difficult to handle. Typical field use involved the labor of more than one person.



AEROSOL GENERATOR FLOW DIAGRAM
Fig. 5.

The early DOP generators were also considered rather messy and perhaps dangerous. Often hot DOP liquid was observed being spewed from the machine in lieu of the desired aerosol during times of inadequate heat transfer and/or aerosol coagulation. These massive generators were prone to suffering damage from shipping and handling as well.

In 1981, the current version of a high capacity (thermal concept) aerosol generator was produced. Named the Model F Aerosol Generator, this machine proved to be a very effective, convenient and safe thermal aerosol generator.

Design and Features

It was the designer's goal, prior to the inception of the Model F generator, to build an instrument with none of the previously mentioned shortcomings. To accomplish this end, the following design features were employed:

- a) The heater block, within which the up to two gram sec⁻¹ liquid DOP is vaporized, was made by casting a 2.5 meter long, helical stainless steel tube within a 1000 cc cylindrical aluminum block, (See Fig. 6). The goal of gaining adequately high heat transfer characteristics was achieved with the long tube, together with the highly heat conductive aluminum block material. The stainless steel tube also offers the advantage of minimal chemical reactivity with the hot DOP.
- b) Adequate thermal input for high capacity applications was gained through the use of several high watt-density cartridge heaters inserted directly into the well insulated heater block (See Fig. 6). As much as 4 kilowatts of energy can be drawn to maintain a stable block temperature above the DOP boiling point throughout the 5-10 minute duration of a typical test. When drawing from common 120 volt sources, two isolated circuits are employed to gain the needed power without overloading any one circuit.
- c) Stable temperature control was accomplished by using Curie point principle magnetic switches. As the heater block reaches its control point temperature, these switches lose their normally magnetic state, subsequently opening the triac controlled heating circuit. Significant space savings over conventional controllers was also realized since the compact switches were mounted directly on the heater block, (See Fig. 6).

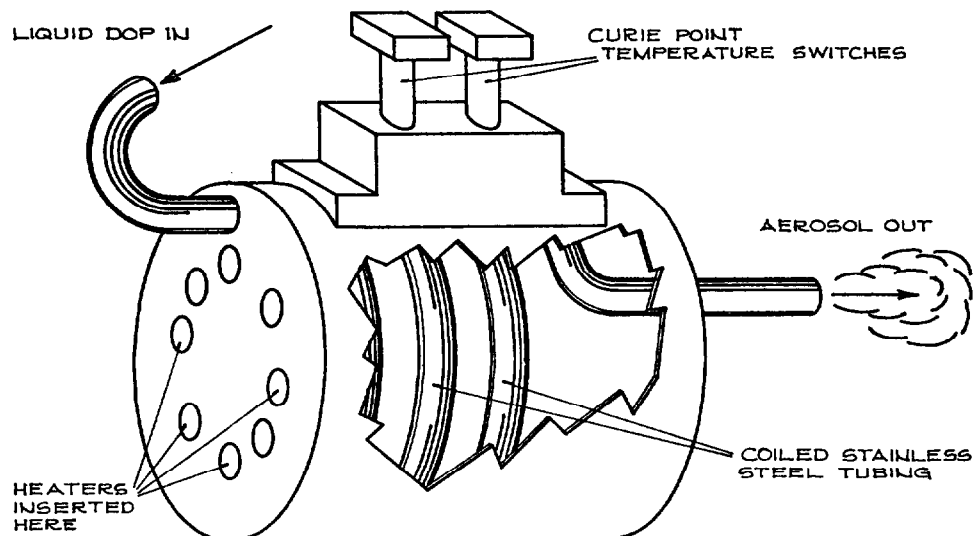


Fig. 6.

AEROSOL GENERATOR
HEATER BLOCK DETAIL

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- d) The generator's weight was minimized by using aluminum to make the compact heater block, resulting in a total instrument mass of less than 20 kilograms. Prudent space utilization made possible a finished instrument volume of less than $.037 \text{ m}^3$ (1.3 ft^3) that is easily handled by one person. Despite the small size, the instrument's 1.4 liter built in fluid reservoir provides enough DOP to operate at its maximum rate for more than 10 minutes.
- e) The problem of safely handling a normally "messy" and hot DOP aerosol was aided by two basic design improvements: First, the aerosol exhaust port was located within the generator's rear enclosure, opposite the control panel. The operator is thus at a distance from the resulting aerosol, and kept clear of any condensing DOP. Secondly, the proper mixing of gas with the liquid DOP, prior to heating in the heater block (See Fig. 5), has been shown to be a very effective method of eliminating the problem of dripping at the generator outlet port.

Gas mixing has also proven to be an effective method of cooling the aerosol more rapidly after exiting the generator than would otherwise occur. The DOP aerosol thus becomes less hazardous in terms of direct contact, and likewise, less flammable.

DOP is generally considered nonflammable. However, when vaporized as is done in the aerosol generator, and mixed with a certain volume of air at a moderately high temperature, the mixture can be considered flammable. Tests performed on operating generators have shown, however, that even given an ignition source, DOP aerosol would not sustain a flame when in the normal operating mode. Reasonable care to avoid open flame in the vicinity of the operating generator is, of course, advised.

Reports to date from air filtration test personnel in the field, have been altogether positive regarding the performance, convenience and safety of this high capacity thermal aerosol generator.

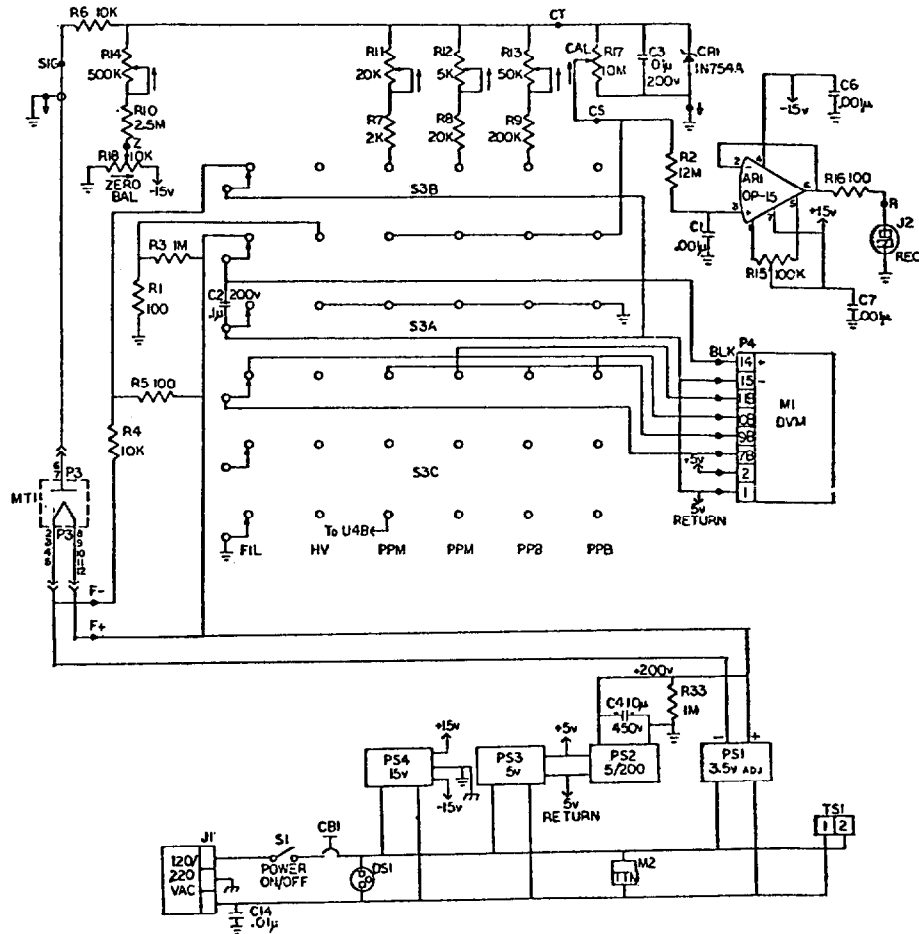
IV. HALIDE GAS DETECTOR

The open diode type sensor has been the standard for leak detection of halides for many years. It offers excellent sensitivity and a wide linear response range. Current fabrication techniques allow millisecond response times and increased life span.

These characteristics provide a solid foundation on which to design a leak detection instrument. In 1974, NUCON designed the first F-1000 Halide Detection System based on an open diode sensor. This system took advantage of these characteristics to provide the first direct reading halide detectors designed for in-place leak testing. Development of the HD-SA Halide Detectors began in 1980. Design goals included increasing the linear range of the detector while improving stability over previous instruments.

Open Diode Sensor

Theory of operation: The heart of the HD-SA Detector is the open diode type sensor. The sensor consists of two platinum electrodes - an anode and a cathode. A DC bias of 200 volts is applied across the two electrodes so that a potential is established. A second DC voltage is used to heat the anode to approximately



HD-SA electrical schematic
Fig. 7.

900°C. A flow of positive ions from the heated anode to the cathode produces a detectable current. This current is proportional to the number and the atomic weight of the halide atoms flowing across the electrodes. Detectable halogens are chlorine, fluorine, iodine and bromine along with common fluorocarbon gases such as the refrigerants R-11, R-12, R-22 and others.

The sensitivity of the element to halides is achieved by specially treating the anode with a ceramic alkali during its manufacture. A monomolecular layer of alkali atoms exists on the surface of the heated anode. When a halide atom contacts this surface, an electron is transferred from the halide atom to the anode. This occurs because the electron work function of the surface is greater than the ionization potential of the gas atom. The halide then removes the alkali atom from the surface of the anode.

The monomolecular layer of alkali atoms is continuously replenished from the ceramic material by migration. Too high a concentration of halides will strip the alkali atoms faster than they can be replenished causing reduced sensitivity of the cell. In effect, the cell reaches a saturation point where its response is no longer linear. This saturation point occurs at approximately 0.5 Vppm for R-11. The cell is also weakly sensitive to oxygen. Since the oxygen background is con-

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stant, it can be easily zeroed out electrically and poses no problem to the detection of halides.

Improved Linear Response Range

Leak testing of carbon filters requires an upstream concentration of 5 Vppm to 10 Vppm of R-11. This concentration, however, exceeds the linear range of the detector element. The sampled gas must be diluted with a known proportion of clear air to bring it within the detection range of the instrument.

The dilution method chosen was to cycle the solenoid valve which controls the TEST/CLEAR inputs to the detector. An electronic circuit was designed to vary the "valve on" time, alternating the sample gas with clear air to extend the dynamic range of the instrument, (See Fig. 8).

The timing pulse of the circuit is based on the 60 Hz line frequency. The timing pulse is sent through a series of decade counters which control the duty cycle of the solenoid. The circuit is "hardwired" programmable for any dilution ratio.

The solenoid is turned on and off by an opto-coupled triac switch. This prevents inductive kickback produced by the solenoid from damaging the timing circuit.

Experimentation on a prototype dilution circuit found that the alternating gas sample and clear air were not mixing well. This caused undesirable fluctuations. A mixing chamber of 250 ml was added upstream to the sample valve. (See Fig. 9) The mixing chamber has an internal baffle which combines the sampled gas with the clear air. This results in a homogeneous sample passing through the sensor element, thereby eliminating the fluctuations and improving the detector readouts. Response range was extended from .5 Vppm to 50 Vppm for R-11.

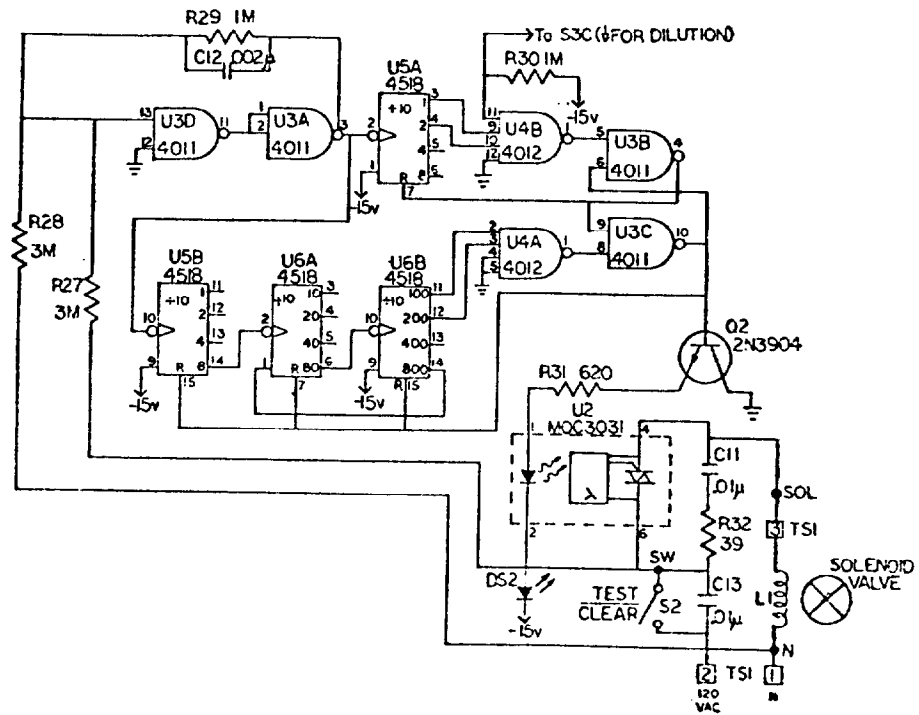
Stability

Short term drift is an excellent indicator of stability. Drift is associated with a constant deviation or fluctuation of an output reading. The greatest amount of drift appears on the maximum sensitivity scale of an instrument. Any minute change in electronics, temperature or flow rates will appear as changes in the readout. Therefore, to increase instrument stability, changes in flow rates, electronics and temperature must be minimized.

Flow Rate Stabilization

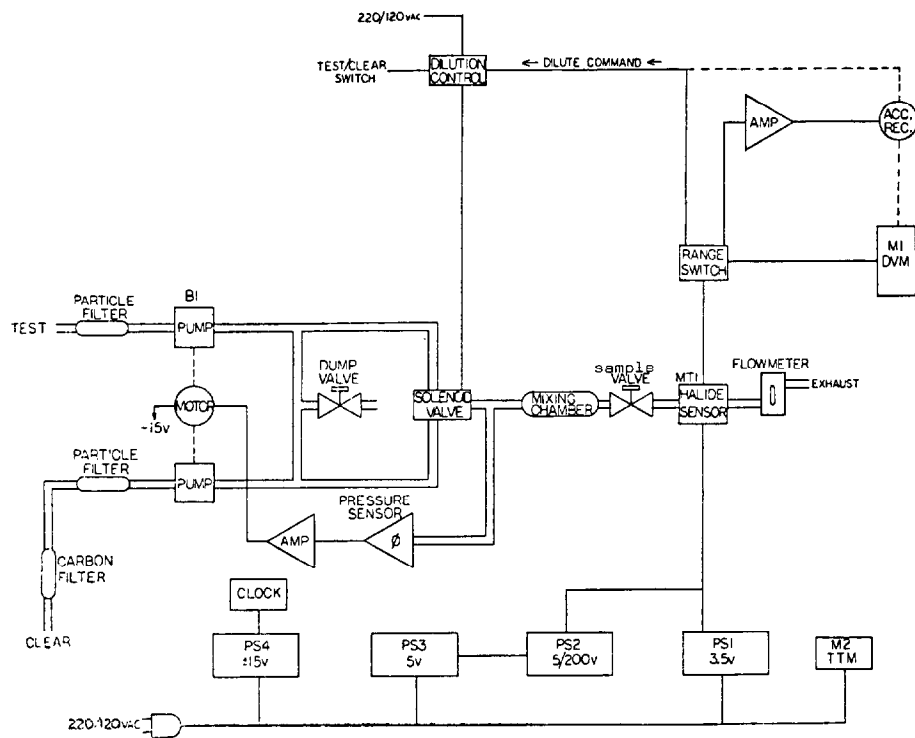
Flow rate stabilization of the pneumatic system is accomplished by using a DC servo-controlled dual diaphragm pump. The pump draws approximately 2.2 l min^{-1} and pressurizes a balanced closed loop system, (See Fig. 9). About 200 ml min^{-1} (approximately 10%) is diverted through the sample valve and then through the sensor. The rest of the air exits through the dump valve at approx. 2 l min^{-1} rate assuring a constant supply of fresh sample gas.

Test of clear air is selected by an electrically operated solenoid valve. The clear air passes through an external carbon filter which removes any background halides. Two 60 micron filters remove any debris which may enter through the sample lines and damage the pump or sensor.



electronic dilution circuit

fig. 8

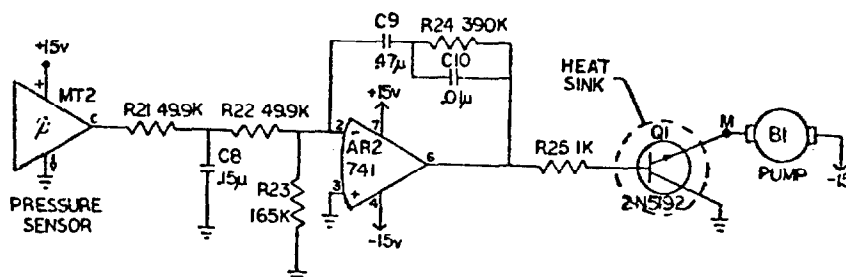


HD-SA flow diagram

fig. 9

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A piezo-resistive type pressure transducer senses any change in the pressurized loop. An electronic circuit then compares the output of the transducer to that of reference voltage. Any change of pressure within the system causes an error signal to be developed. This error signal changes the pump motor speed, which corrects the imbalance in the loop. (See Fig. 10).



electronic flow regulator circuit

fig. 10

Early NUCON detectors used a pneumatic feedback regulator to stabilize the flow across the sensor. The pneumatic regulator proved to be slow and insensitive in responding to rapid change and tended to drift from set flow rates.

The electronic feedback regulator has the advantage of being quick to respond to changes. It will keep a steady flow across the sensor with ± 1 kPa (± 4 " H₂O) pressure changes on the sample intake. This assures an even flow across the sensor reducing the drift of the electronic signal.

Electronic Stability

Stability of the electronics is achieved by using power supplies that are actively regulated to $\pm 0.01\%$ and exclusive use of reliable and accurate solid state components. The digital readout meter is qualified per MIL-STD-202 for accuracy, ruggedness and temperature stability.

Temperature Stability

Improved temperature stability of the instrument was achieved through the design of a special sensor housing. This housing is machined from aluminum and has a mass of 400 grams. The block acts as a heat sink for the sensor cell and provides thermal capacitance to ambient temperature changes. Temperature stabilization occurs after a warm up period of about 30 minutes.

A series of channels drilled through the interior of the housing allows gases to be pre-heated before flowing into the sensor cell section of the housing. Pre-heating of gases and the resistance to temperature changes help to control short term drift and, therefore, improving stability of the instrument.

V. HALIDE GAS GENERATOR

The ability to perform reliable leak detection of activated charcoal filter systems with a fluorocarbon (otherwise called "halide") gas requires the use of not

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only a sensitive, stable detector as explained in the previous section, but also a repeatable, stable source of the challenge gas. This section of the paper explains the rationale behind the design, and describes the NUCON Model F Halide Gas Generator.

The most prominent fluorocarbon gases used are the common refrigerants Fluorotrichloromethane, (R-11) and Dichlorodifluoromethane (R-12). Boiling points for R-11 and R-12 are 23°C and -30°C respectively at one atmosphere. Thus, each must be kept at elevated pressures to maintain a liquid state. R-11 and R-12 are ideal challenge agents due to their ready vaporization and low carbon adsorption characteristics. However, specialized equipment is necessary to provide convenient handling of these fluorocarbons in field test applications.

The current version of a high capacity fluorocarbon gas generator was developed in 1981. Designated the Model F, Halide Generator, this instrument proved to be simple to operate, safe, and rugged field test tool. It was designed and built to provide on-site test personnel with a very portable instrument capable of delivering a known quantity of challenge gas in sufficient volumes to reliably test filter systems of any size currently in use.

Design and Features

A series of earlier generator models made clear the need for an instrument that successfully met the following four parameters:

- a) Adequate capacity to test $38 \text{ m}^3 \text{ sec}^{-1}$ (80,000 cfm) filter systems.
- b) A convenient, portable, "field ready" design.
- c) Operator safety.
- d) Challenge gas output stability.

To meet the need for adequate capacity to test large filter systems, it was considered necessary to provide at least 10 minutes of challenge gas generation at a constant 10 Vppm upstream concentration. A 1.4 liter capacity fluorocarbon pressure reservoir was built into the Model F to achieve this requirement.

A total instrument volume of 1.3 cu. ft. achieved the needed portability and convenience. Controls for the instrument consist of four valves mounted on a front panel. The fluid reservoir was placed within the enclosure resulting in a single-unit instrument (unlike earlier dual unit models), yet total weight was kept to 12 kilograms.

Some early NUCON generator models were designed to be filled by pouring the R-11 or R-12 into an open top container. In this way, the reservoir fill level could be observed, but it proved inconvenient and wasteful since much of the liquid boiled away. The Model F, however, is filled through a "charging port" located on the control panel, and reservoir fill is monitored with a panel mounted sight column. Filling is thus more convenient, and a less wasteful operation. It also prevents the release of halide during filling which would result in high background concentrations for tests performed in small enclosed areas.

The improved filling method greatly enhances operator safety as well, since fluorocarbon concentrations are kept low during filling. R-11 and R-12 gases are relatively harmless, however, the OSHA Permissible Exposure Level requires that personnel not be subject to inhaled concentrations greater than 1000 Vppm. This criterion is met by the proper use of the Model F generator.

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Finally, a most important parameter for a functional generator is the capability of providing a repeatable and stable source of challenge gas. Experiments performed on earlier models revealed this to be a shortcoming that had to be corrected.

The main problem of past models was inherent in gas-metering designs. The inevitable cooling and gas density variations had a proportional effect on output concentration. This problem was solved by mixing air with the fluorocarbon after metering, but before exiting the instrument. Figure 11 depicts the basic flow

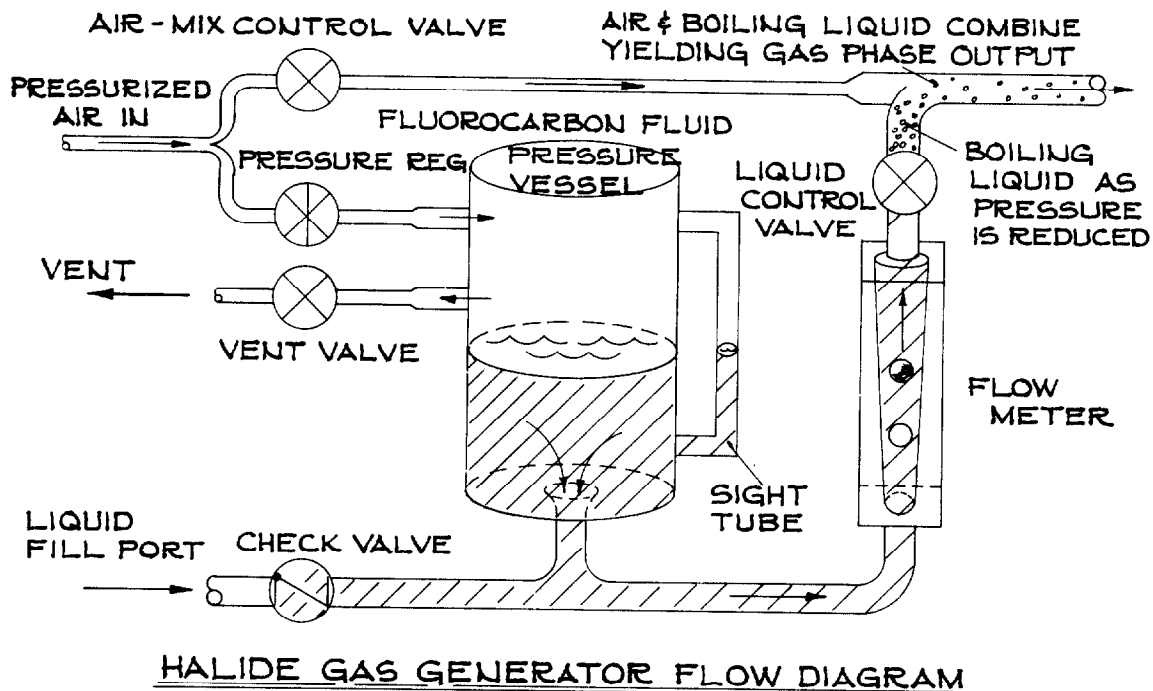


Fig. 11.

configuration used with the Model F, and shows where this air/liquid mixing occurs. As long as a stable elevated pressure exists within the liquid circuit, a simple flowmeter yields excellent metering stability and repeatability. A wide metering range is accomplished through the use of dual floats of unequal density.

In summary, the Model F Halide Generator demonstrates many advantages over earlier designs in terms of capacity, convenience, safety and stable output. Together with the F-1000 Halide Detector, truly accurate quantitative leak testing of filter systems even as large as $38 \text{ m}^3 \text{ sec}^{-1}$ (80,000 cfm) is easily accomplished.

CONCLUSION

The design and operation of a complete family of nuclear air filtration system test instruments developed for both field and laboratory use is described. The instrumentation has been extensively field tested and found to exceed the requirements. The capabilities of the NUCON F-1000 filter leak test instrumentation are in the realm of the next generation of air cleaning systems where leaks less than 0.01% must be detected.

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ACKNOWLEDGEMENT

The authors wish to thank the group of NUCON field test engineers for suggestions and numerous tests performed during the design of the instrumentation.

Thanks to be given also to Mr. C. E. Graves, S. L. Homonnay and C. L. Fry for their assistance with the assembly of this report.

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Fig. 12.

NUCON F-1000 Field Test Leak Detection System

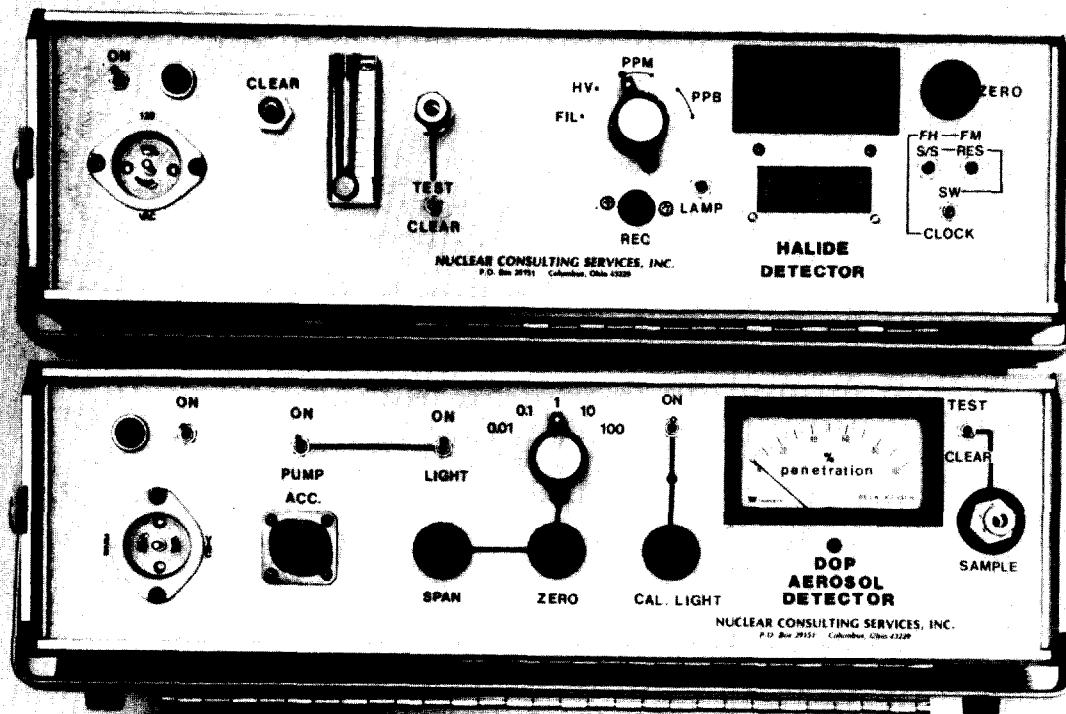


Fig. 13.

NUCON Gas and Aerosol Detector

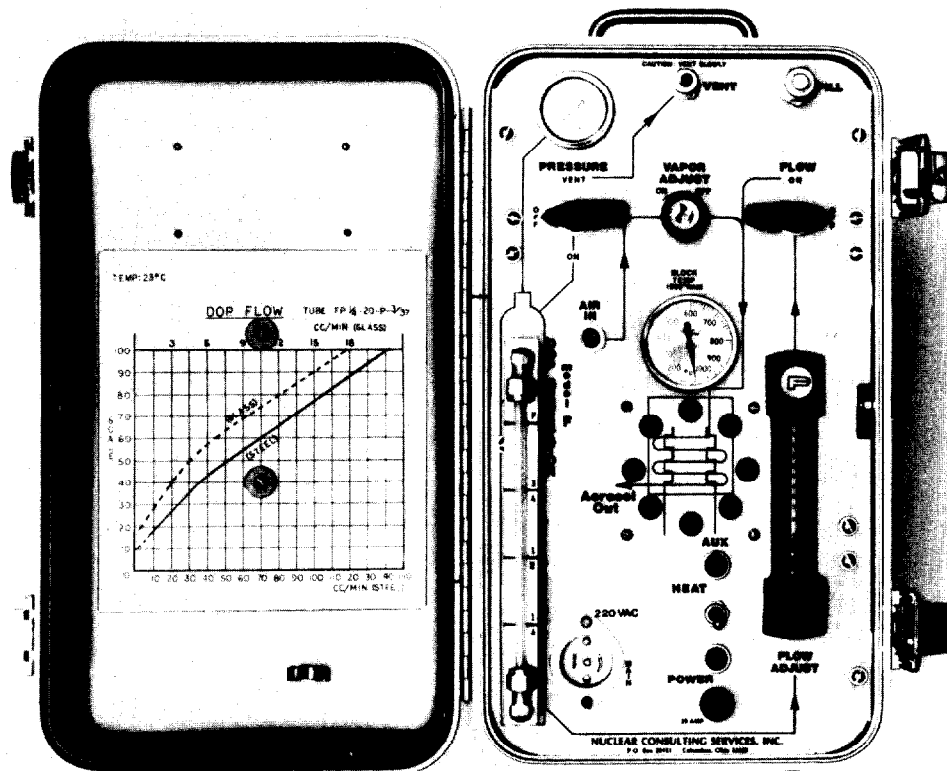


Fig. 14.

NUCON DOP Aerosol Generator

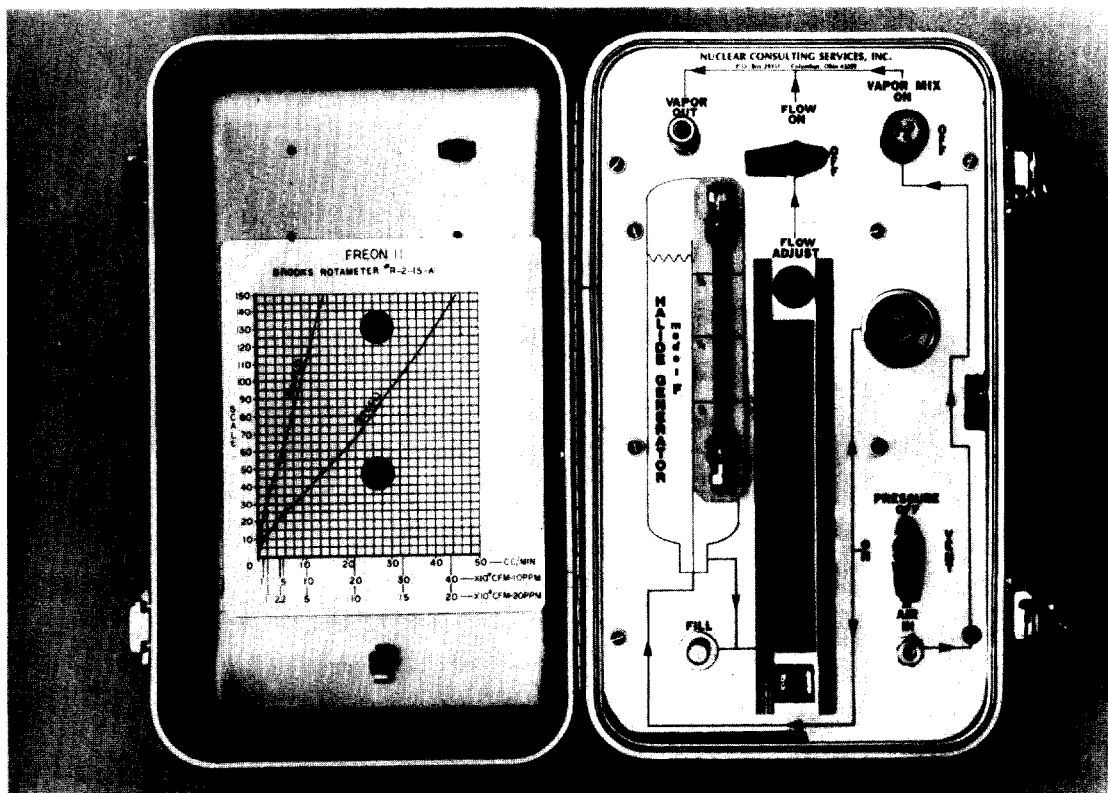


Fig. 15.

NUCON Halide Gas Generator

DISCUSSION

ANDERSON, W.L.: Since the theory shows that the amount of light scattered is proportional to the inverse fourth power of the wave length, why did you design the instrument to operate in the infra-red rather than the untraviolet wavelength range?

KOVACH, J.L.: Our design did not eliminate the ultraviolet (UV) part of the spectrum, but extended the used spectrum in both directions. The previously used incandescent lamp works at 3000°K with maximum intensity at 900 nm (near infra-red) whereas the ultraviolet emission was absorbed in the regular lime glass envelope. Our quartz-halogen lamp works at 3400°K with a peak emission at 800 nm. It has an extended emission characteristic up to 200 nm (into the UV range) which is not absorbed because the quartz envelope is transparent to the UV emission. Our light detector has an extended range up to 200 nm, as well, but it is also sensitive down to 1200 nm in the infra-red region. Although the previous photomultiplier covered the spectrum from 300 to 600 nm (See Figure 2 in the paper), it used only a small fraction of the emitted light. Our lamp-detector combination is matched to use most of the useful spectrum, including more UV than was previously used.

ANDERSON, W.L.: Have you considered the possible explosive hazard resulting from using air as the gaseous propellant for your high capacity generator?

KOVACH, J.L.: Yes. It has been proven experimentally that a $\frac{1}{4}$ inch (4 mm inside diameter) stainless steel tube acts as a flame arrestor, even for highly flammable propane gas. Consequently, neither explosion nor flame can travel or develop inside the tubing between the DOP reservoir and the exit port. After the mixture exists, it is mixed with a large amount of air, so the influence of the relatively small amount of propellant gas becomes unimportant, because the mixture falls below the lower explosive limit a short distance from the exit.

POLYMERIC DIFFUSION AS APPLIED TO
A RADIOIODINE OFF-GAS MONITOR

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Abstract

Potentially, the most significant airborne wastes from nuclear power reactors are radioactive iodine isotopes. Large beds of charcoal, silver zeolite, or other silver-impregnated materials are used to isolate these radioiodines from the environment. Yet, verification of continued operation of the bed as originally designed is important from both safety and regulatory viewpoints.

Although sensitive detection methods for ^{131}I exist, interferences from radioxenons and radiokryptons require rapid, on-line separations. If these separations could be achieved, the simultaneous measurement of the radioiodine concentrations upstream and downstream of the adsorbent bed could supply a continuous verification of bed performance.

This paper reports the use of selective diffusion through polymeric membranes to perform these separations. Iodine separations from ^{85}Kr have been demonstrated in fuel reprocessing plant stack gases.⁽¹⁾ Reactor off-gases require larger separation factors than demonstrated in fuel reprocessing plant stack gases (10^6 versus 10^4) and separation from radioxenon.

The work was performed in three phases: 1) permeation constants of the membrane material for iodine compounds and noble gases were measured; 2) a membrane device was constructed that would separate the iodine compounds from noble gases; and 3) the integrated monitor was demonstrated with noble gas and iodine mixtures.

Three conclusions were drawn from these evaluations: 1) adequate separation factors ($>10^6$) can be achieved by combining pulse height analysis with the selective permeation separations; 2) sufficient selectivity exists to reliably measure cleanup filters with efficiencies greater than 99%; and 3) the efficiency measurement is insensitive to changes in off-gas composition.

Introduction

The ability of some polymers to separate permanent gases has been known for many years.⁽²⁾ The law governing the permeation of a gaseous constituent across a membrane is:

$$J = \frac{P\Delta p}{h} \quad (1)$$

where: J is the flux of a gaseous constituent across the membrane;

P is the permeability of the polymer to the diffusing gas;

Δp is the difference in bulk partial pressure of the diffusing gas across the membrane;

h is the thickness of the membrane.

From Equation 1 it can be inferred that the flux of a gas through a given membrane can be predicted if the membrane's permeability is known. Conversely, if the permeability of several gases (I_2 , CH_3I , Xe, Kr) are known, their separation factors can be derived for a given membrane.

Using the equations derived by Treece, et al.⁽³⁾, these factors can be derived for a tube and shell membrane configuration (Figure 1), a configuration easily adapted to continuous off-gas monitoring. If we define K (the separation factor) as the ratio of the partial pressure of the diffusing gas in the product to the partial pressure of the diffusing gas in the purge (see Figure 1), then:

$$K = \frac{\Phi \ln(b/a)}{2\pi LP p_t} + 1 \quad (2)$$

where: Φ is the volumetric flowrate of the purge stream;
 L is the length of the polymeric tubing;
 p_t is the total pressure of the product stream;
 b is the outer diameter of the tubing;
 a is the inner diameter of the tubing.

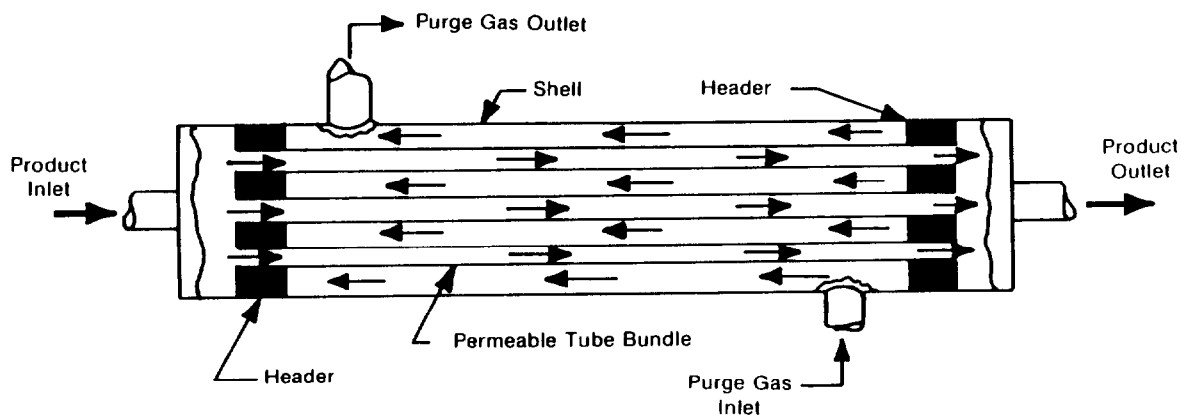


Figure 1

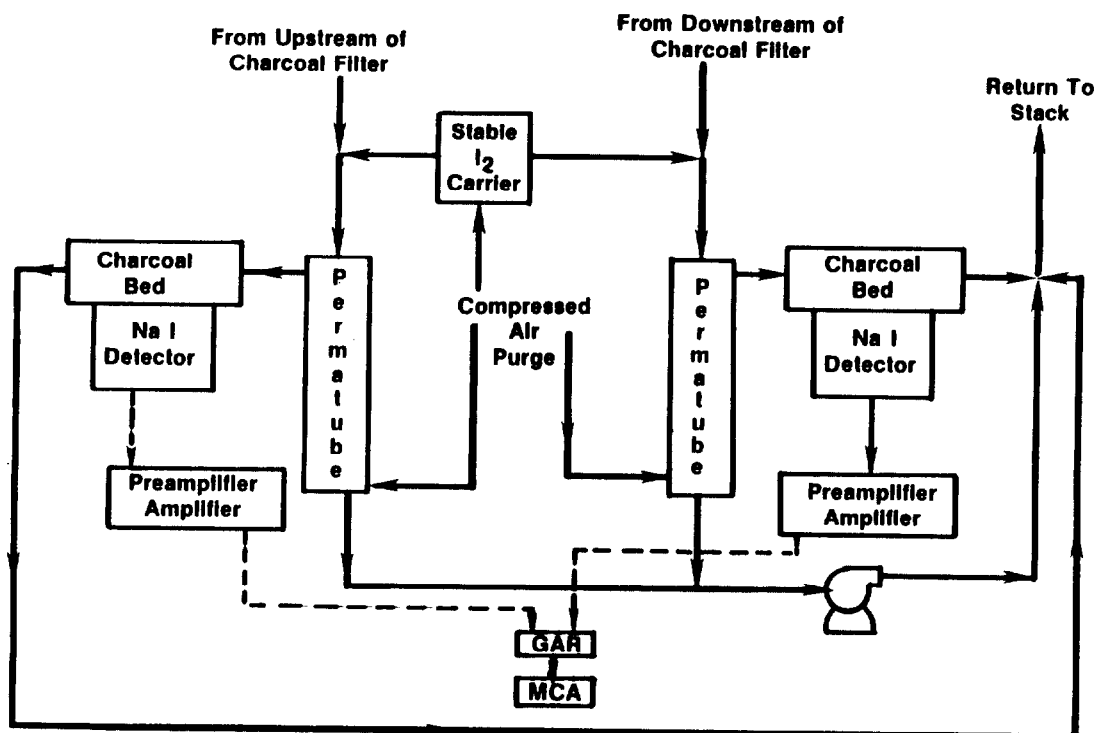
Schematic Diagram of a Permeation Device

From Equation 2, permeabilities (and thereby separation factors) can be calculated if the partial pressures in the purge and product streams are measured. Conversely, once the permeabilities are known, Equation 2 can be used to design a device to achieve the desired separation factors.

The objective of the work presented in this paper was to develop a device that, based on these principles would separate ^{131}I from the radioactive isotopes of xenon and krypton. If suitable reactor off-gas separation factors (on the order of 10^6) could be achieved (either by physical separation or gamma spectroscopy), then a radioiodine cleanup filter efficiency monitor could be constructed. A conceptual diagram of this monitor is shown in Figure 2. (The perm tubes in Figure 2 contain the polymeric membranes in the tube-and-shell configuration shown in Figure 1.) This monitor would simultaneously measure the ^{131}I concentration upstream and downstream of a charcoal or silver zeolite cleanup filter thereby providing continuous verification of filter integrity.

Phase I - Determination of Permeation Constants

During the first phase, experiments were performed to measure the permeability constants of CH_3I , I_2 , Kr and Xe. This information was then used in designing a perm tube that would achieve the desired separation factors. The apparatus to measure the I_2 and CH_3I permeation constants is shown in Figure 3. A $900 \text{ cm}^3/\text{min}$ methane flow was mixed



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Figure 2

INTEGRATED MONITOR

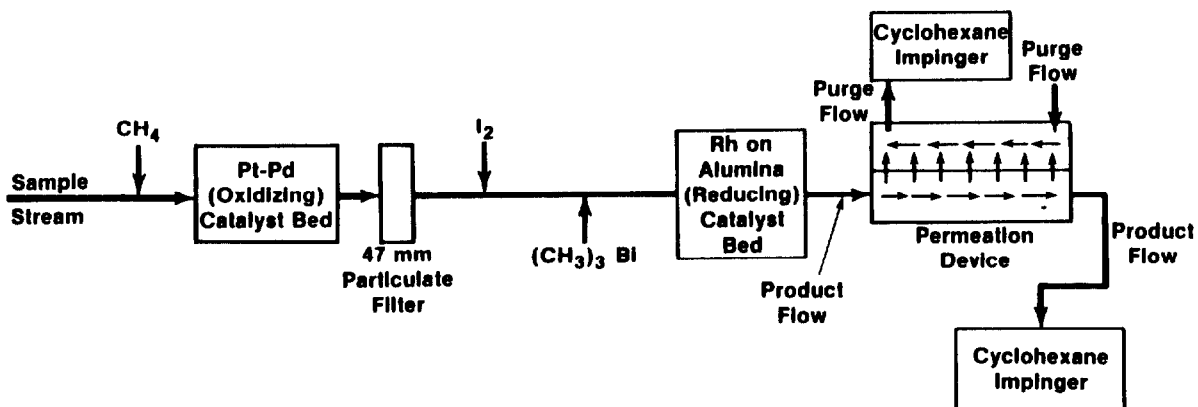


Figure 3

IODINE COMPOUND EXPERIMENTAL APPARATUS

with a $90 \text{ cm}^3/\text{min}$ air flow. This air - methane mixture was then passed over a 4 g Pt-Pd catalyst bed at 500°C to convert the O_2 in the sample stream to CO_2 . After filtering the sample stream with a 47 mm Gelman Type A/E glass fiber filter, sufficient $(\text{CH}_3)_3\text{Bi}$ and stable I_2 were added to make the final concentration $15 \text{ mg I}_2/\text{m}^3$ and 0.5 mg Bi/m^3 . This $990 \text{ cm}^3/\text{min}$ flow then passed over the Rh on alumina catalyst bed (maintained at 500°C where a fraction (typically 40-70%) of the I_2 was converted to CH_3I . Subsequently, the product flow was directed into the permeation device containing 317.5 cm of Silastic tubing. The purge flow was maintained at 2.83 L/min . Both the purge and product flows were bubbled through 50 mL of distilled-in-glass grade cyclohexane, and the CH_3I and I_2 analyzed by gas chromatography using electron capture detection.

The results of the iodine compound experiments are shown in Table I. Using Equation 2 with these constants indicates that a 317 cm long permeation tube maintained at 2.0 L/min purge flow and 1.0 L/min product flow would separate 37% of the I_2 and 55% of the CH_3I into the purge stream.

TABLE I
RESULTS OF IODINE PERMEATION EXPERIMENTS

<u>Compound</u>	<u>Number of Determinations</u>	<u>Permeability ($\text{cm}^3 \text{ (STP)}/(\text{cm} \cdot \text{cm Hg})$)</u>
I_2	7	3×10^{-5}
CH_3I	8	5×10^{-5}

Next, the permeability constants of Kr and Xe were determined. These measurements required the apparatus shown in Figure 4. Using mass flow controllers, the sample flowrate was maintained at $100 \text{ cm}^3/\text{min}$ and the purge flowrate at 1.4 L/min . The ^{85}Kr and ^{133}Xe tracers were injected upstream of a 500 cm^3 mixing chamber. The product inlet concentration and purge outlet concentration of ^{85}Kr or ^{133}Xe were then measured with 2-L ionization chambers. Two HP 3390A reporting integrators processed the signals from the two 2-L ionization chambers.

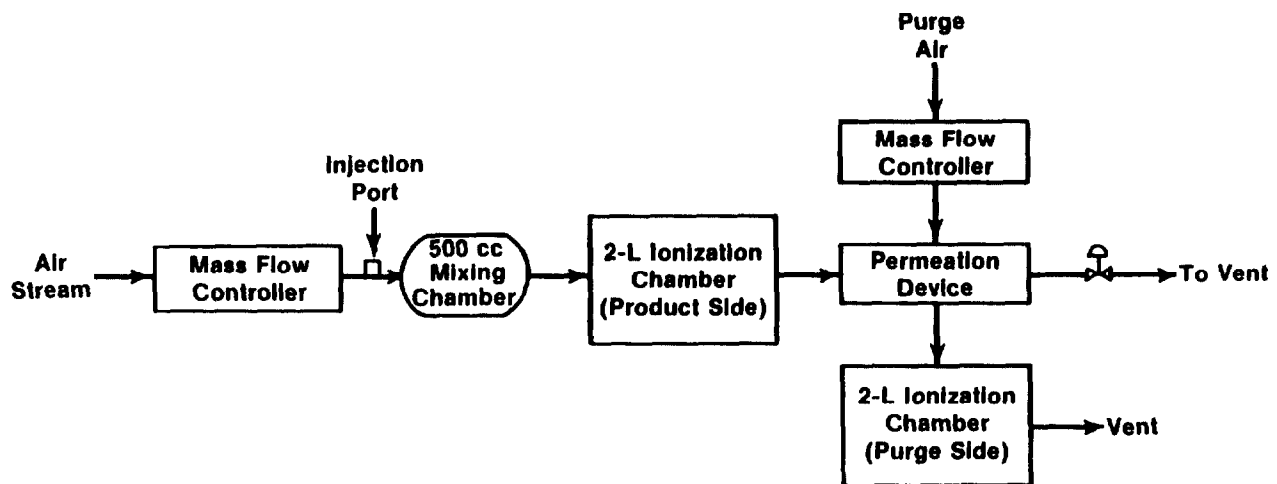


Figure 4

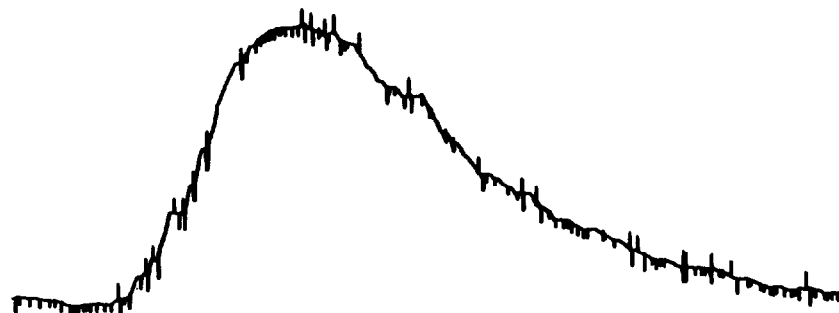
KRYPTON AND XENON PERMEATION APPARATUS

The results of two typical krypton and xenon tests are shown in Figures 5 and 6. Figure 5 is the response of the two ionization chambers to an injection of ^{85}Kr . The purge side ionization chamber was ten times more sensitive than the product side ionization chamber during this test. Therefore, the purge side response represents quantitative transport of the ^{85}Kr across the membrane. This would represent an effective permeability for ^{85}Kr of $5 \times 10^{-3} \text{ cm}^3 (\text{STP})/(\text{cm} \cdot \text{cm Hg})$, four orders of magnitude greater than predicted from the measured permeabilities of other permanent gases in Silastic tubing. One possible explanation is the migration of ^{85}Kr by exchange with the stable Kr dissolved in the membrane. The effective permeability can be used to calculate separation factors, although this experiment emphasized the necessity of confirming separation factors with radioisotopes.

The same isotopic exchange was not observed with ^{133}Xe . Figure 6 shows the result of an experiment identical to that just noted but using ^{133}Xe . A typical ^{85}Kr purge response is included for comparison. An effective permeability of $3 \times 10^{-9} \text{ cm}^3 (\text{STP})/(\text{cm} \cdot \text{cm Hg})$ was calculated from these experiments. One possible reason the ^{133}Xe does not migrate across the membrane is the stable Xe concentration in air is 13 times less than the Kr concentration. Therefore, the concentration of Xe atoms available for exchange would be lower by at least an order of magnitude.



Product Side Injection Of ^{85}Kr



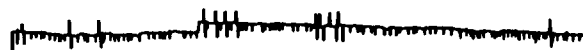
Purge Side Response To Product Side Injection Of ^{85}Kr

Figure 5

Typical ^{85}Kr Test



^{133}Xe Product Injection



^{133}Xe Purge Response



^{85}Kr Purge Response To Calibration Injection

Figure 6

Typical Xe Test

Phase 2 - Membrane Device Construction

The separation factors shown in Table II can be calculated for a 317 cm perm tube constructed of Silastic tubing and operated at a purge flow of 2L/min and a product flow of 1L/min. As shown in Table II, a separation factor of $\sim 10^4$ for radioiodine from Xe can be achieved with the perm tube alone. Therefore, if an additional discrimination factor of 100 can be achieved by pulse height analysis, the separation factors shown in Table II will be satisfactory, i.e., satisfy the separation factors required for reactor off-gases. Therefore, a 317 cm Silastic perm tube was used in phase 3: demonstration of the integrated monitor.

TABLE II

PREDICTED SEPARATION FACTORS

	<u>From CH₃I</u>	<u>From Kr</u>	<u>From Xe</u>
	3		
I ₂	1	1	1×10^4
CH ₃ I	1	1	1.6×10^4
Kr	1	1	4×10^5
Xe	1.6×10^4	4×10^5	1

Phase 3 - Demonstration of the Integrated Monitor

The integrated monitor test apparatus shown in Figure 7 was constructed to determine the actual achievable separation factors. The noble gases were injected with a 10cc gas tight syringe. The $^{125}\text{I}_2$ permeation source was a 1/4" x 1/2" x 2" sealed Silastic tube containing 50-100 mg of I₂ crystals that had been traced with ^{125}I . The permeation rate was 30 $\mu\text{g I}_2/\text{min}$ and 3 pCi $^{125}\text{I}/\text{min}$.

The iodine was separated from the noble gases in a 317 cm perm tube maintained at a purge flow of 1 L/min. The charcoal bed collected the separated I₂ and the ^{125}I was counted with a one inch by one milli-

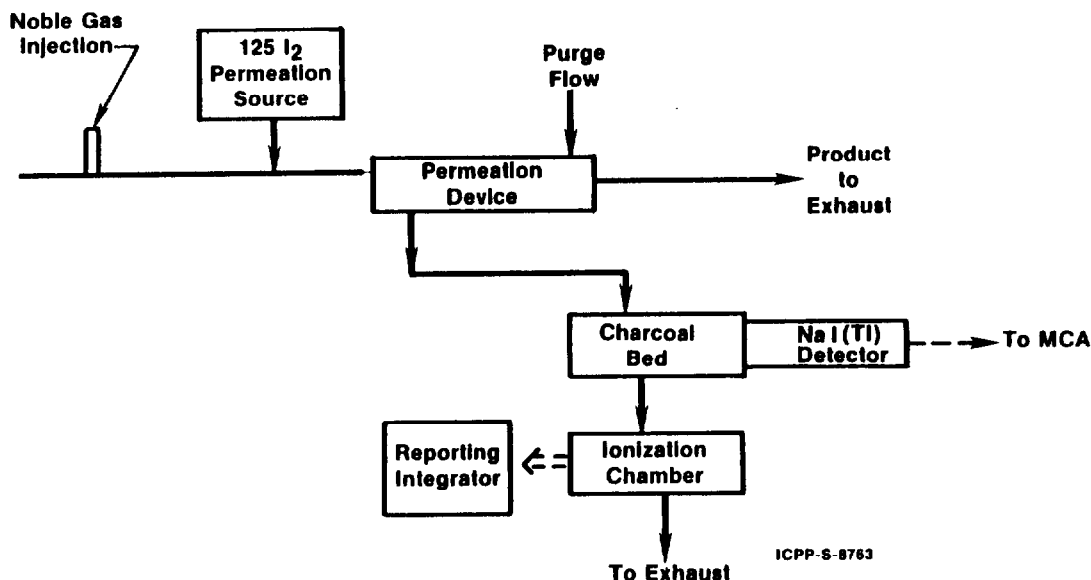


Figure 7

INTEGRATED MONITOR TEST APPARATUS

meter NaI(Tl) (FIDLER) detector. The signal from the FIDLER was processed by an ND60 multichannel analyzer.

A ^{125}I energy region was selected that overlapped the 80 keV ^{133}Xe gamma ray but did not overlap the 514 keV ^{85}Kr gamma ray. Therefore, the observed ^{133}Xe separation factor would represent the separation factor achieved with selective permeation alone; the ^{85}Kr separation factor represents the separation factor achieved instrumentally by pulse height analysis. The 2-L ionization chamber measured the noble gases actually penetrating the permeation device.

The results of this experiment are plotted in Figure 8. The monitor's response to ^{125}I of 10^8 cps $(\mu\text{Ci/cc})^{-1}\text{d}^{-1}$ remained constant over the duration of the experiment. After removal of the $^{125}\text{I}_2$ permeation source, a memory effect persisted for several days. This memory effect was about 5% of the ^{125}I signal in the presence of the $^{125}\text{I}_2$ permeation source.

The cause of this memory effect is unknown, but there are two possible explanations. First, the collected I_2 may migrate upward within the charcoal bed and toward the detector. This migration would then

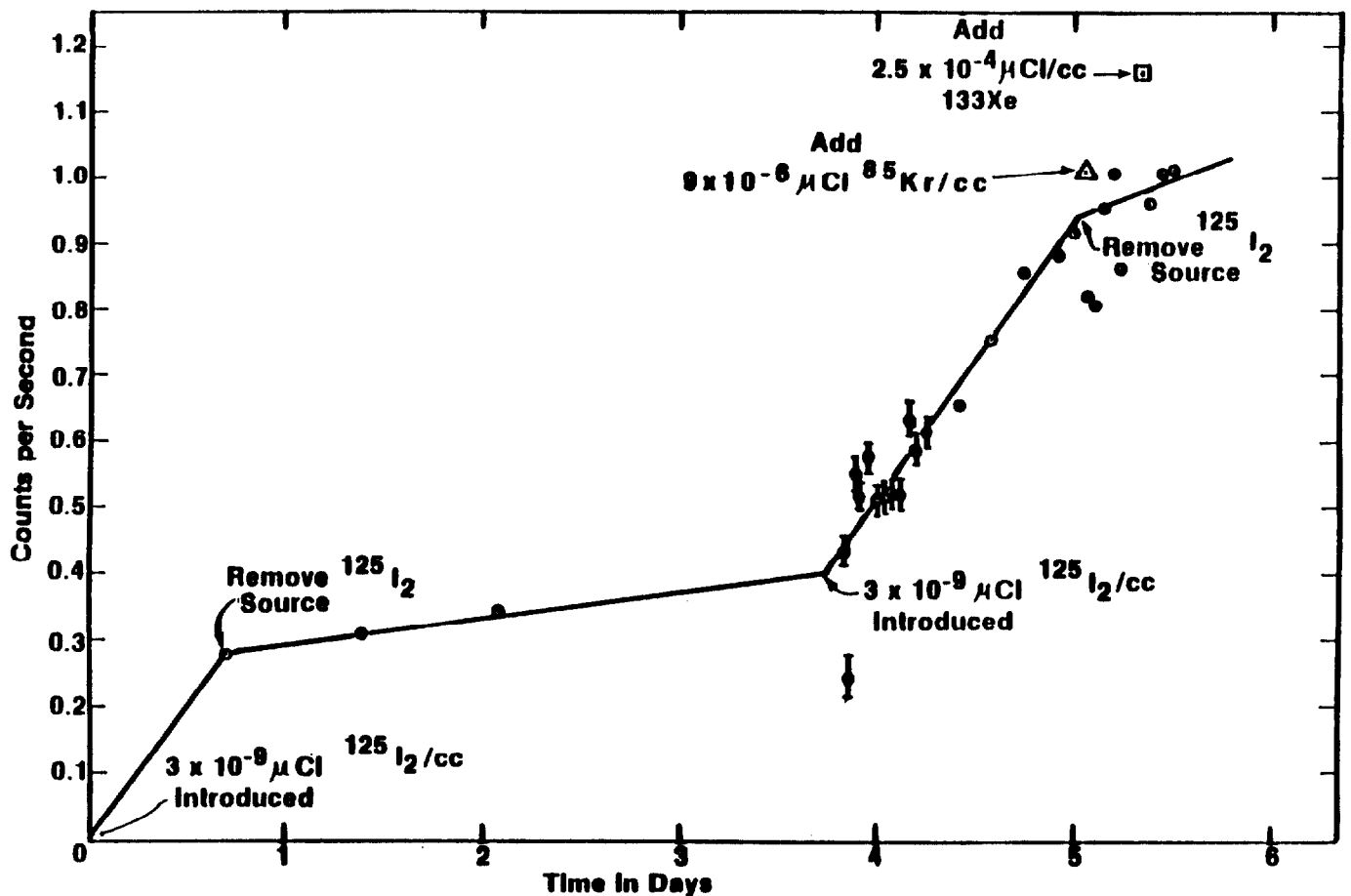


Figure 8

IODINE, KRYPTON AND XENON MIXTURE EXPERIMENT

change the counting geometry. It is unlikely, however, that the migration rate within the charcoal bed would remain constant over periods as long as several days. The second explanation is that residual $^{125}\text{I}_2$ that had dissolved in the polymeric tubing was continually permeating into the purge stream after the $^{125}\text{I}_2$ source was removed. If residual $^{125}\text{I}_2$ in the polymeric tubing was the cause of the memory effect, then conversion of the I_2 to CH_3I by previously published techniques⁽⁴⁾ may resolve this problem.

As shown by the immediate increase in count rate after reinsertion of the I_2 permeation source, the response time of the permuted system is less than 15 minutes.

During the day 4 to day 5 time period, H_2 was used instead of air as the product carrier gas. Owing to its anomalous permeation behavior,

hydrogen was selected as a worst case carrier gas to demonstrate the effect of changes in off-gas composition. The fact that no change in monitor response was observed demonstrated the unit was insensitive to changes in off-gas composition.

The response of the integrated monitor to a 3×10^3 $^{85}\text{Kr}/^{125}\text{I}$ ratio and a 8.6×10^4 $^{133}\text{Xe}/^{125}\text{I}$ ratio is also shown in Figure 8. The measured separation factors are compared to the predicted separation factors in Table III. These data indicate that the permeation device achieved separation factors equal to or greater than predicted.

TABLE III

SEPARATION FACTORS ACTUALLY ACHIEVED

<u>Isotope</u>	<u>Predicted Separation Factor</u>	<u>Observed Separation Factor</u>
^{133}Xe	1.5×10^4	2.4×10^4
^{85}Kr	100	900

In addition, the data plotted in Figure 8 indicate that there was no discernable memory effect due to the presence of noble gases. Therefore, the permeation device used in this experiment may have application to a radioiodine cleanup filter efficiency monitor.

Conclusions

The major conclusions of this work were the following:

- 1) Separation factors greater than 10^6 for xenon and krypton can be achieved;
- 2) Response times less than 15 minutes are possible;
- 3) Memory effects for elemental radioiodine on the order of a few percent of the average radioiodine concentrations were observed;

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- 4) The monitor response is insensitive to off-gas composition;
- 5) Radioxenon and radiokrypton cause no discernable memory effects.

Accordingly, selective permeation may have application to a radioiodine cleanup filter efficiency monitor.

References

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2. J. Crank and G. S. Park, Diffusion in Polymers, Academic Press, London, 1968.
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4. S. J. Fernandez and B. G. Motes, "A Continuous Realtime Radioiodine Monitor Employing On-Line Methyl Iodide Conversion", paper presented at the 180th National Meeting of the American Chemical Society, Las Vegas, NV, August 24-29, 1980.

DISCUSSION

GILBERT, What are the prospects for development of practical hardware for the industry?

FERNANDEZ: Funding is pending. We have a mandate to develop the instrument and to encourage its implementation but there are other organizations within Exxon Nuclear responsible for funding and the funding level has been indicated.

MURROW: Are the permeation units up-and downstream identical and do they have to be calibrated periodically?

FERNANDEZ: We don't have any trouble with detector sensitivity, so that we try to match the two permeation devices as closely as possible, i.e., as closely as we can manufacture them. Although they are identical, correction factors have to be used. The permeation constants for every elastic tube such as we use, change somewhat depending upon how much the tubes are stretched or stressed. Therefore, whenever we make one of these tubes, there is always a slightly different amount of stressing or stretching that takes place and we have to calibrate each tube.

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SCHOLTEN: Two questions: First, why not use a ceramic adsorber in the detectors, such as AC 6120? Then you have no adsorption, of noble gases and consequently, no disturbance. Second, in a working reactor, the disturbance caused by other iodine isotopes, such as I^{132} , I^{133} , I^{134} and I^{135} , gives a much greater response to your detector than I^{131} , as I explained at the CEC seminar at Mol last year. Have you overlooked that problem?

FERNANDEZ: To answer your question, the reason we did not use silver-impregnated materials for collection is because silver and zeolites and silver-containing silica gels have a much higher adsorption for iodine, so we chose charcoal to increase sensitivity to carbon-14. The answer to your second question is that it is something we have to look at in plant demonstrations.

CLOSING REMARKS OF SESSION CHAIRMAN:

Hanford and Exxon are trying to introduce new systems into their filter plenums to utilize equipment that is installed in-place, thereby making it unnecessary to put people inside the plenums, as we used to do. I am very happy to see that because at Rocky Flats, as an example, where we have many, many systems, we still are checking the HEPA filters manually. But after seeing the data presented by Exxon and Hanford, I will be installing additional piping in some of our plenums to see how it works. Ray forgot to mention that he used PVC piping in one of the areas. With DOP, it very rapidly fell apart. So I won't be using PVC. Another area that we will be looking at, is the use of 1,800 and 1,000 CFM filters. I don't know how many people will realize how important this is to the industry until they get into the design and construction of new facilities. If we can come up with good pre-filtration and larger capacity HEPA filters, the amount of money that can be saved in the construction and operation of facilities will be tremendous. With larger capacity filters, we can reduce systems to almost half the size. Presently, we use 500 or 1,000 CFM for the design of filtering systems. With 1,800 CFM filters, we could save on waste disposal as well as on construction. I would like to see more work done on prefilters to determine the optimum size and efficiency that should be used for prefilters to extend the life of HEPA filters. At present, we use 80 - 95% roughing filters throughout Rocky Flats and various other types of roughing filters are used throughout the industry. I find that they help extend the life of HEPA filters and I would like to learn what is the best roughing filter efficiency for protecting the life of HEPA filters. I highly recommend them for modification and renovations, as well as for installation of new systems. We have found at Rocky Flats, and I am sure at other installations as well, that now that we are modifying our systems because of new criteria established for all of the facilities throughout the United States, and throughout the world, that when we install new systems we immediately tear up the old HEPA filtering systems. So I would like to know more about the roughing filters.

I think that it is too bad that the work at Harvard is going to be terminated because of lack of funding because I don't think the data are definitive or the results conclusive. I doubt that the correct prefilter efficiency was employed. Perhaps a different prefilter would have made a difference in the plugging rate of the HEPA filters. Dr. Kovach has described new portable instruments with unique features that we should take a look at to see if they offer advantages for carbon and filter testing. Mr. Fernandez's work appears to have real potential and I hope that funding will come so that he can produce hard data to confirm preliminary results.